Master's thesis

NTNU
Norwegian University of Science and Technology
Faculty of Natural Sciences
Department of Biology

Connor McKnight

Establishing the food web structure and mercury concentrations in an Arctic coastal lagoon

Master's thesis in Environmental Toxicology and Chemistry
Supervisor: Bjørn Munro Jenssen
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Abstract

Arctic coastal lagoons are dynamic ecosystems which make up large portions of the Arctic coastline. Despite this, lagoon ecosystems have seldom been studied in the Arctic, therefore there is limited knowledge on food web structures and contaminant concentrations in these ecosystems.

Mercury (Hg) is a non-essential, toxic heavy metal which has previously been shown to accumulate in Arctic environments at enhanced concentrations that can induce toxic effects in high trophic level species. Due to lagoons being heavily influenced by the surrounding terrestrial environment, lagoons may be potential hotspots for Hg accumulation as climate change is leading to increased melting and transportation of terrestrially derived organic matter and associated contaminants such as Hg. We therefore developed this study with the intent to provide baseline information on a coastal lagoon on Svalbard (Richardlaguna) including data on food web structure and concentrations of total mercury (TotHg) and methylmercury (MeHg) in water, sediments and biotic samples.

The lagoon food web (based on available samples) was comprised of brown macroalgae, littoral amphipods, polychaetes, bivalves, gastropods, holothurians, priapulids, shorthorn sculpin (*Myoxocephalus scorpius*) and Arctic staghorn sculpin (*Gymnocanthus tricuspis*). Stable isotopes of carbon ($\delta^{13}C$) revealed that lagoon fauna generally relied on marine carbon sources compared to terrestrial carbon sources. Values of stable nitrogen isotopes ($\delta^{15}N$) were used to approximate the trophic level of lagoon fauna, which revealed a trophic range of 1-4.2 (brown macroalgae-shorthorn sculpin).

Lagoon stream inlets had the higher concentrations of aqueous and particulate Hg than the lagoon and the outer marine environment, which highlights the importance of terrestrial inputs as sources of Hg to lagoon ecosystems. Concentrations of TotHg differed significantly between particulate organic matter and sculpin (p=0.0001), as well as macroalgae and sculpin (p=0.003), with a range of 0.68-418 ng/ g dw. Based on these data, we calculated a trophic magnification slope of 0.18 and a trophic magnification factor of 3.4, which indicated that TotHg is biomagnifying through the lagoon ecosystem.

These findings highlight the importance of Arctic coastal lagoon ecosystems as potential hotspots for inputs of terrestrially derived organic matter and Hg. To the authors knowledge, this is the first study to report stable isotope data and environmental concentrations of Hg in a lagoon ecosystem from Svalbard.

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Trondheim, May 2019,

Connor McKnight

Contents

Abbreviations

AMAP	Arctic Monitoring and Assessment Programme
BMF	Biomagnification factor
Chl *a*	Chlorophyll a
cm	Centimetres
cm^2	Centimetres squared
CRM	Certified reference materials
CTD	Conductivity temperature depth
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
dw	Dry weight
EPA	Environmental Protection Agency
g	Grams
GF/F	Glass fibre filter
ha	Hectares
HCl	Hydrochloric acid
H_2SO_4	Sulphuric acid
Hg	Mercury
Hg0	Elemental mercury
Hg^{2+}	Mercuric cation
k	Condition factor
km	Kilometres
L	Litres
Log_{10}	Log_{10} transformed data
m	Metres
m^2	Metres squared
mDOM	Marine derived organic matter
MeHg	Methylmercury
mg	Milligrams
mm	Millimetres

mV	Millivolts
n	Number of observations
ng/g	Nanograms per gram
ng/L	Nanograms per litre
NH_4^+	Ammonia
NIVA	Norwegian Institute for Water Research
NO^{2-}/NO^{3-}	Nitrate/Nitrite
NTNU	Norwegian University of Science and Technology
NTU	Nephelometric turbidity unit
OM	Organic matter
p	Significance level
PE	Polyethylene
PMeHg	Particulate methylmercury
PO_4^{3-}	Phosphate
POC	Particulate organic carbon
POM	Particulate organic matter
ppt	Parts per thousand
PTotHg	Particulate total mercury
QMA	Quartz fibre filter
R^2	R-squared
SIA	Stable isotope analysis
SiO_4	Silicate
Sn	Tin
SPM	Suspended particulate matter
$SUVA_{254}$	Specific ultraviolet absorbance at 254 nanometres
tDOM	Terrestrially derived organic matter
TL	Trophic level
TMF	Trophic magnification factor
TMS	Trophic magnification slope
TN	Total nitrogen

TOC	Total organic carbon
TotHg	Total mercury
TP	Total phosphorous
UNIS	University Centre in Svalbard
ww	Wet weight
WoRMS	World Register of Marine Species
µg/L	Micrograms per litre
µm	Micrometres
$\delta^{13}C$	Ratio of stable carbon isotopes ($^{13}C/^{12}C$)
$\delta^{15}N$	Ratio of stable nitrogen isotopes ($^{15}N/^{14}N$)
$\Delta^{15}N$	Average enrichment of $\delta^{15}N$
°C	Degrees Celsius
%MeHg	Percentage of mercury present as methylmercury

1. Introduction

1.1. Contaminants in the Arctic

The Arctic is currently undergoing rapid environmental change, including warming, changes in biodiversity, socio-economic challenges and the long-range transport of pollutants (Callaghan et al, 2004; Berkman & Young, 2009; Bennett et al, 2015). The combined effects of these environmental stressors on the Arctic environment generates a high degree of uncertainty in terms of the future environmental health of this region.

The occurrence of anthropogenic contaminants in the Arctic, including persistent organic pollutants and heavy metals such as mercury (Hg) is concerning given that this region is generally viewed as being pristine. Although local point sources of pollution are few, elevated contaminant concentrations have been consistently observed in both the abiotic environment and in biota. The phenomenon of enhanced contaminant concentrations in the Arctic is attributed to long range transport from southerly latitudes. Many pollutants (including Hg) can be transported to and throughout the Arctic through atmospheric and oceanic currents, drifting sea ice, biological vectors and the large Arctic rivers (Barrie et al, 1992; Burkow & Kallenborn, 2000; Blais et al, 2005; Verreault et al, 2010). Atmospheric transport from southern regions is likely the most important transport route for contaminants to the Arctic. This is due to "global distillation" (Fernández & Grimalt, 2003; O'Driscoll et al, 2005), the process through which semi-volatile contaminants are released to the atmosphere at lower (and warmer) latitudes and transported to the Arctic, where colder conditions lead to condensation and deposition. Reduced re-volatilisation due to lower temperatures can lead to accumulation of contaminants and therefore enhanced environmental concentrations. The Arctic is therefore described as a sink for chemicals which possess this nature (Ariya et al, 2004; AMAP, 2016).

1.2. Mercury

Mercury (Hg) is a non-essential heavy metal and a potent pollutant due to its bioaccumulative, toxic and persistent properties (Clarkson & Magos, 2006; Liu et al, 2011; Scheuhammer et al, 2015). Although Hg occurs naturally in the environment via sources such as volcanic eruptions and erosion of rocks (e.g. cinnabar), environmental concentrations often greatly exceed the natural background level (Andersson et al, 2008; Dietz et al, 2013; Soerensen et al, 2016). This is due to anthropogenic activity and industrialisation, with Hg emissions linked to activities such as the burning of fossil fuels, waste incineration, mining

activities (e.g. artisanal and small-scale gold production) and metal production (ferrous and non-ferrous) (AMAP, 2011).

As atmospheric Hg (Hg0) is deposited due to the cold conditions in the Arctic, transformation to other Hg species such as ionic Hg (Hg^{2+}) can occur through biogeochemical processes and microbial activity. This allows for movement between different compartments of the environment (Chetelat & Braune, 2012). Organic matter (OM) is particularly important for the mobilisation of Hg as Hg^{2+} readily binds to OM by forming bonds with sulphur containing groups (e.g. thiols) present on the OM molecule (Skyllberg et al, 2000; Haitzer et al, 2003). After bonding to OM, Hg can be transported between different environmental compartments (e.g. export from terrestrial to aquatic ecosystems). However, bottom sediments often act as the ultimate sink for Hg once OM particles settle down to the benthic environment. Through the action of sulphate reducing bacteria (which are present in bottom sediments), Hg is transformed into organic forms such as methylmercury (MeHg) which readily enters food webs and bioaccumulates. The bioaccumulative properties of MeHg are attributable to its affinity for binding to proteinaceous tissues, especially those with sulphur containing groups (e.g. thiols) and structures associated with the amino acid cysteine (Harris et al, 2003; Clarkson & Magos, 2006). MeHg also efficiently biomagnifies through food webs, leading to enhanced concentrations in biota at high trophic levels including marine mammals, seabirds and predatory fish (e.g. Basu et al, 2009; Beyer & Meador, 2011; Chetelat & Braune, 2012; Dietz et al, 2013; Tartu et al, 2013; Krey et al, 2015; Scheuhammer et al, 2015). Aquatic food webs are particularly vulnerable to the effects of biomagnifying contaminants like MeHg, as they often have longer food chains compared to terrestrial systems (Gray, 2002).

Organic Hg compounds have the highest toxicity potential and are potent neurotoxins, being able to cross the blood-brain barrier (Clarkson & Magos, 2006). Various health effects have been observed in wildlife including mammals, birds and fish including neurotoxicity (e.g. memory loss, ataxia, paresthesia, tremors) , teratogenic effects (smaller offspring, deformed embryos) and reproductive toxicity (unsuccessful hatching of eggs, decreased egg production, reduced fertilisation) (Liu et al, 2011; Mastromonaco, 2016). Ultimately, MeHg can cause death at the highest accumulated concentrations. Similar effects have also been observed in humans, with historically severe MeHg poisoning events (which led to mortality) including the mass poisoning of residents from Minimata Bay, Japan (1959) and the grain disaster of

Iraq (1971). It is therefore imperative to monitor concentrations of Hg (including MeHg) in the environment, so that potential effects associated with toxicity can be negated.

1.2.1 Mercury in the Arctic environment

It has been estimated that 80-140 tonnes of Hg are accumulated in Arctic food chains each year, with most of this being present as MeHg (AMAP, 2011). Many studies have reported Hg levels which exceed toxicity thresholds for several Arctic species (Basu et al, 2009; Dietz et al, 2013; Tartu et al, 2013; Scheuhammer et al, 2015). For example, cellular damage (necrosis) and hepatic fibrosis has been observed in freshwater fish species e.g. Arctic char (*Salvelinus alpinus*) (Drevnick, 2012; Drevnick, 2013). For marine fish, Hg concentrations in Greenland shark (*Somniosus microcephalus*) have been shown to be at concentrations which can induce changes to biochemical processes, damages to cells and tissues and detrimentally impact reproduction (Beyer & Meador, 2011; Chetelat & Braune, 2012). Concentrations of Hg in shorthorn sculpin (*Myoxocephalus scorpius*) from West Greenland were reported to have Hg concentrations capable of causing reproductive toxicity through reduced reproductive performance (Sonne et al, 2014). Neurotoxicity in the form of neurochemical and neurobehavioral changes has been reported in several species of marine mammal e.g. ringed seals (*Phoca hispida*), beluga whales (*Delphinapterus leucas*), and polar bears (*Ursus maritimus*) (Krey et al, 2015). Reproductive toxicity has been reported in black-legged kittiwakes (*Rissa tridactyla*) from Svalbard, which were shown to exhibit abnormal reproductive hormone responses and skip breeding when exposed to elevated Hg concentrations (Basu et al, 2009; Tartu et al, 2013). This highlights the potential for Hg to generate a broad range of toxic effects, which range from cellular levels up to population level effects in Arctic systems. Particular attention should be payed to lower trophic species, as knowledge of levels at the base of food chains and biomagnification of Hg in Arctic food webs are important for assessing potential toxic effects in higher trophic level organisms.

Humans inhabiting Arctic environments such as Inuit communities are also susceptible to heightened Hg concentrations. This is due to traditional practices which involve the hunting and consumption of high trophic level food sources (e.g. marine mammals), which act as a vector for human exposure (Bjerregaard et al, 2004; AMAP, 2011). Although there has been a general decline in blood Hg concentrations in humans of Arctic communities, many people exceed the guideline levels for Hg in blood (AMAP, 2011). People who exceed the guideline levels are mostly represented by vulnerable groups such as pregnant women and their unborn children (Chetelat and Braune, 2012). A study from Nunavut showed that 60% of pre-school

children were actively exceeding the weekly tolerable intake of MeHg according to limits set from the World Health Organisation (Tian et al, 2012). Current Hg levels in inhabitants of some northern communities are at levels which could potentially induce toxic effects. However, dietary advice is proving to be a useful strategy for controlling the quantity of Hg received from consuming traditional foods (AMAP, 2011).

1.3. Arctic coastal environments

Arctic coastal environments are areas representing the link between the terrestrial and marine environments. These regions are currently in a state of environmental transition, as climate change is leading to increased melting of snow, ice, glaciers and permafrost. This is causing alterations in precipitation patterns, which can lead to increased rates of erosion and delivery of freshwater to coastal environments (Dunton et al, 2006; Dunton et al, 2012; Harris et al, 2017; Harris et al, 2018). These changes are driving an increase in loading of terrestrial materials from land to sea, including increased inputs of freshwater, sediments, nutrients, OM and contaminants such as Hg, all of which have potential implications on coastal biogeochemistry, hydrography, ecology and contaminant cycling. Terrestrial inputs also act as a direct transport vector for various contaminants (including Hg), thus meaning that coastal regions may act as potentially important Hg accumulation zones (Outridge et al, 2008; Zhang et al, 2015).

1.3.1 Lagoon ecosystems

Lagoons are shallow bodies of water which are found on all continents, making up 13% of the global coastline (Barnes, 1980; Kjerfve, 1994). Within the Arctic, lagoons represent approximately a third of the Arctic coastline (Haynes & Robards, 2017). These features occur inland and have a close orientation towards the coastline and are therefore considered to be transitional ecosystems between land and sea (Pérez-Ruzafa et al, 2011). These systems may be completely closed to the open marine environment or be semi-isolated. Those which are semi-isolated possess a barrier with an opening that allows for active exchange with the ocean, hence they are referred to as open coastal lagoons (Barnes, 1980). Coastal lagoons (including those found in the Arctic) are some of the most productive ecosystems in the world (Knoppers, 1994; Dunton et al, 2006; Duck & da Silva, 2012; Dunton et al, 2012) and provide important ecosystem services e.g. providing food sources for northern communities (Harris et al, 2017). These habitats show high annual variability in salinity and temperature, which is even more pronounced in the Arctic due to this region's extreme seasonality (Harris et al, 2017). Some Arctic regions like the Beaufort Sea coastline showing temperature ranges

of -2 to 14°C and salinity ranges of 0 to >45 (Harris et al, 2017). Organisms inhabiting these environments must therefore be able to tolerate environmental extremes.

Coastal lagoons are inhabited by a wide range of organisms. Diverse assemblages of benthic fauna have been recorded in Arctic coastal lagoons, which include dense populations of marine invertebrates such as polychaetes, gastropods, bivalves, crustaceans, ascidians and sponges (Dunton et al, 2012). These benthic species employ a range of feeding strategies (e.g. deposit feeding, filter feeding, scavenging and active predation) (Dunton et al, 2006; Macdonald et al, 2010) and some organisms like polychaetes are important and preferred prey items for higher trophic level consumers, including fish and seabirds (Brown et al, 2012). Common fish species found in Arctic coastal lagoons include Arctic char (*Salvelinus alpinus*), Arctic flounder (*Plueronectes glacialis*), Arctic cod (*Boreogadus saida*) and sculpins (*Myoxocephalus spp*.) (Craig et al, 1984). Numerous avian species also utilise coastal lagoons and it is thought that >150 migratory bird species (including waterfowl) (Brown, 2006) utilise these habitats (plus other coastal environments) as feeding grounds during the summer months (Churchwell et al, 2016). Previous observations also noted beluga (*Delphinapterus leucas*) and bowhead whales (*Balaena mysticetus*) actively foraging in the open waters surrounding lagoons along the Beaufort coastline (Pedersen & Linn, 2005).

Like other coastal environments, lagoons are highly influenced by terrestrial inputs. The heightened influx of terrestrial materials to lagoons and reduced exchange with open marine waters compared to other coastal systems may make these environments potential sinks for terrestrial carbon, nutrients and Hg (Naidu et al, 2003; Misra et al, 2006). The increased loading of terrestrially derived organic matter (tDOM) could potentially be an important source of energy for lagoon fauna, as previous studies in Arctic coastal lagoons have indicated that tDOM is an important subsidy for marine derived organic matter (mDOM) (Dunton et al, 2012; Harris et al, 2017). Aquatic microbes can use tDOM as an energy source, which in turn can be an important food source to primary consumers (e.g. zooplankton) and can cause a shift in diet from phytoplankton to microbial food sources (Berggren et al, 2015; Karlsson et al, 2015; Tanentzap et al, 2017). Although, a shift to a microbial based diet is of less quality due to microbes having a lower quantity of essential fatty acids in comparison to phytoplankton (Arts et al, 2009). This change in energy source may have implications in environments receiving high concentrations of tDOM (like Arctic coastal lagoons) in terms of food web structure, quality of food at the base of the food web and trophic efficiency, which can in turn impact the biomagnification of contaminants like Hg which can bind to OM.

Recent findings from a study on boreal lakes from Norway, showed that tDOM can influence the MeHg concentration of aquatic biota. Findings suggested that increasing concentrations of tDOM led to higher concentrations of MeHg in water and zooplankton by directly increasing aqueous concentrations of MeHg and indirectly by altering MeHg bioavailability and changing the flow of energy in the lower food web (Poste et al, 2019). Arctic coastal lagoons may therefore be important accumulation zones of Hg and consequentially may expose biota to elevated exposure of Hg through tDOM and lead to biomagnification of this contaminant in Arctic coastal food webs.

1.4. Uses of stable isotopes in food web studies

The fundamental uses of stable isotope analysis (SIA) in ecological studies are based on two widely accepted assumptions: 1) stable isotope values reflect an organisms diet 2) there are natural differences in stable isotope ratios which can be used to distinguish food sources and different habitats (Jardine et al, 2006). By using values of stable carbon isotopes ($\delta^{13}C$) and stable nitrogen isotopes ($\delta^{15}N$), it is possible to establish the origin of OM in ecosystems (e.g. marine vs. terrestrial) and determine the relative trophic level (TL) of organisms respectively (Hobson & Welch, 1992; Jardine et al, 2003). Estimations of TL are based on the sequential enrichment of ^{15}N ($\Delta^{15}N$) upon progression up the food chain (Hobson & Welch, 1992), were the heavier ^{15}N isotope is retained within consumers while the lighter ^{14}N isotope is excreted more readily.

Previous studies have utilised SIA to determine the relative importance of terrestrial vs. marine carbon sources in Arctic coastal lagoons (Dunton et al, 2006; Dunton et al, 2012; Harris et al, 2018). The application of SIA is also widely used in ecotoxicology studies, to study the exposure of contaminants via diet and to assess the biomagnification of contaminants like Hg in food webs (Jardine et al, 2006). Past studies from a range of Arctic ecosystems have shown positive correlations between TL (derived from $\delta^{15}N$) and Hg concentrations (e.g. Atwell et al, 1998, Rigét et al, 2007; Jæger et al, 2009; Gantner et al, 2010; Clayden et al, 2015) which also report the biomagnification of Hg based on this relationship.

1.5. Aims and objectives

Studies on Arctic coastal lagoon ecosystems are sparse, despite their high density along the Arctic coastline, ecological importance and sensitivity to climate change. In particular, very

little is known regarding contaminant concentrations and contaminant cycling in these ecosystems, and almost all the information available on Arctic coastal lagoons is from studies based in the North American Arctic. Recently, the Norwegian Polar Institute identified over 100 coastal lagoons (>5ha in size) in Svalbard, which vary in morphology, degree of connectivity to the coastal marine environment (e.g. closed, open with low exchange, open with high exchange), and catchment land-cover (e.g. glaciers, permafrost, bedrock) (Haug & Myhre, 2016). To our knowledge, the current study is the first to report detailed environmental data from a coastal lagoon on Svalbard. This project was also designed to be a pioneering study, with the main objective being to report baseline Hg concentrations in abiotic and biotic compartments of a coastal lagoon in order to give insight into Hg contamination and cycling in these systems.

The main aims of this project were to:

1) Investigate the food web structure of a coastal lagoon system on Svalbard (Richardlaguna) and determine the relative importance of terrestrial vs. marine energy sources for lagoon fauna
2) Determine the concentrations of Hg (TotHg and MeHg) in water, sediments and biota from the lagoon environment
3) Determine if Hg is biomagnifying through the lagoon food web

2. Materials & methods

2.1. Study location

All sampling was conducted at Richardlaguna (78°46'N, 10°57E) in early September (1st-3rd) 2018. Richardlaguna is a coastal lagoon which is located on the north-east of Prins Karls Forsland (see Figure 2.1.). It is a narrow lagoon (2-3km wide) with an elongated shape that has an area of 750ha (Haug & Myhre, 2016). The terrain directly surrounding the lagoon is relatively flat. Moving inland from the lagoon, the topography becomes more varied, with plateaus in the south and north, and mountains plus river valleys to the west (Johansen & Overrein, 2011). Several landward ponds are also present (Johansen & Overrein, 2011), with some of their associated streams actively feeding into the lagoon.

Figure 2.1. Aerial map of Svalbard, showing Richardlaguna (78°46'N, 10°57E) as denoted by the red circle. Map courtesy of the Norwegian Polar Institute. Map constructed and taken from: https://toposvalbard.npolar.no

Richardlaguna is home to a wide array of marine fauna. Previous observations have noted the presence of several pinniped and seabird species, as well as Arctic char (Haug & Myhre, 2016). The lagoon habitat is important for both native and migratory fauna. For example, the sand banks act as one of the largest haul-out sites for walrus (*Odobenus rosmarus*) from the months of May-early August (Lydersen & Kovacs, 2014). Black-legged kittiwakes have also been observed resting and bathing at the lagoon, with aggregations comprising hundreds of individuals (Kempf & Sittler, 1988). For these reasons, the lagoon often attracts tourist boats throughout the summer months.

2.2. Field sampling

Field work was carried out as part of a broader field campaign related to the TerrACE project, a Norwegian Research Council project focusing on biogeochemical and ecological effects of terrestrial inputs on Arctic coastal ecosystems. Samples were collected at six areas of the lagoon environment (Figure 2.2.), which included two stream inlets, the lagoon central basin, a nearshore station to deploy gillnets, the lagoon outlet and an outer marine station.

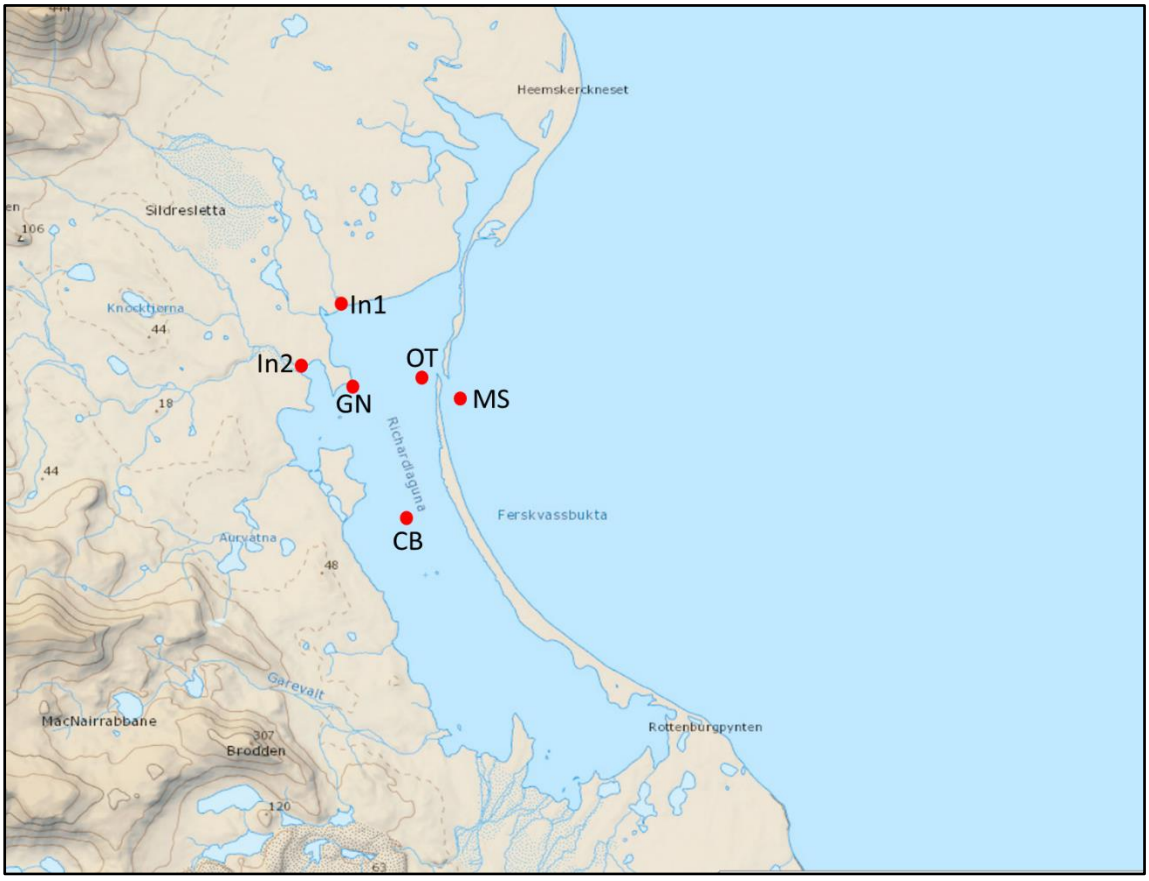

Figure 2.2. Aerial map of Richardlaguna which depicts all sampling locations (red dots): nearshore stream inlets (In1, In2), gillnet station (GN), lagoon central basin (CB), lagoon outlet (OT) and outer marine station (MS). Map courtesy of the Norwegian Polar Institute, taken from: https://toposvalbard.npolar.no

2.2.1 Physical parameters

In order to characterise the physical setting of the lagoon (i.e. temperature, salinity, turbidity), CTD casts were conducted using a handheld Seabird Electronics CTD. For the stream inlets, casts were done by orientating the CTD horizontally and submersing it in the centre of the stream for approximately one minute. For the remaining 'deeper' stations, depth was measured using an acoustic handheld depth sensor. CTD casts for these sites measured the full length of the water column, down to ~0.5m above the sediment/water interface. This was done to avoid contact with the benthic environment, as resuspended material can interfere with the CTD sensors. In addition, Secchi depth was measured at the deepest station (CB) to provide an estimate of water transparency.

2.2.2 Water sampling

Surface water samples (~1m) were collected for analysis of total organic carbon (TOC) and total nitrogen (TN), total phosphorous (TP), TotHg, MeHg, dissolved nutrients (NH_4^+, NO^{2-}/NO^{3-}, PO_4^{3-}, SiO_4) and optical characterisation of dissolved organic matter (DOM) at 254nm ($SUVA_{254}$). This was done for all sampling stations, except for the site were the gillnets were deployed. Samples for TOC, TN, TP, TotHg and MeHg were collected using a pre-cleaned stainless-steel bucket. These samples were immediately transferred to sterile sample bottles (either amber glass or plastic bottles), with Teflon lined caps. For dissolved nutrients, dissolved organic carbon (DOC) and DOM characterisation, an addition filtration step was needed, therefore surface water was collected using a pre-washed 10L carboy and stored in cold and dark conditions until further processing (see section 2.3.1). Prior to collection, all sample bottles (except for DOM characterisation samples) were pre-loaded with 1mL of a preservation agent (either H_2SO_4 or HCl) and double bagged in polyethylene (PE) bags to prevent sample contamination. Further detail on water sampling (e.g. bottle type, sample volume, preservation agents, storage conditions) is displayed in Appendix A. Finally, salinity (ppt), temperature (°C) and pH were determined using a handheld multi-sensor (Hanna Instruments 18195), while turbidity (NTU) was measured using a turbidity sensor (Thermo Scientific Eutech TN-100).

2.2.3 Sediment sampling

Surface sediment samples (top 1-2cm) were collected from two grabs using a $0.025m^2$ Van veen grab at the lagoon central basin (CB). Upon recovery, the grab was checked to ensure that the metal jaws were closed. If there was any indication that the grab was open, the

sample was discarded. For successful grabs, a portion of the surface sediments were transferred to PE bags using a stainless-steel spoon. The temperature (°C), pH and redox potential (mV) of the sediment was also determined using a handheld pH/mV sensor (Hanna Instruments HI9125).

2.2.4 Biological sampling

A range of biological samples, (including brown macroalgae, amphipods, benthic invertebrates and fish) were opportunistically collected at various lagoon locations (see Figure 2.1). Macroalgae (*n*=5) was collected either by hand opportunistically at the shoreline or using gillnets. Care was taken to only select 'fresh' looking samples, and those which had complete structures (e.g. stipe and frond present). Similarly, amphipods (*n*=384) were also collected opportunistically by hand, through turning over rocks. Benthic infaunal samples were collected using a 0.025m^2 Van veen grab (*n*=10 grabs) at the lagoon central basin. For benthic infauna, grab contents were transferred into a large plastic tray for washing to rid the samples of sediments. Once all sediment was removed, organisms from all grabs was transferred to a plastic bucket filled with lagoon surface water (*n*=15 species collected). For sampling of fish (*n*=18; 2 species), gillnets were deployed in a nearshore area of the lagoon and left overnight. Upon retrieval, fish were separated based on species and then immediately double wrapped in aluminium foil to prevent contamination. All biotic samples were then frozen at -20°C.

2.3. Sample preparation

All water samples were stored in cool (4°C) and dark conditions except for TotHg and MeHg samples, which were kept frozen at -20°C with all biotic and sediment samples. Biological samples were identified to the lowest possible taxonomic level based on literature, taxonomic keys and online resources e.g. the World Register of Marine Species (WoRMS). Once classified, benthic fauna were separated into separate pooled samples. The wet weight (ww), length range and number of individuals (*n*) were also determined for pooled amphipod and benthic samples (Appendix B). Trace metal clean techniques were used for laboratory work involving the handling of samples for Hg analysis and water chemistry.

2.3.1 Filtration

Filtration was done at the University Centre in Svalbard (UNIS) filtration lab. Prior to processing, filters were pre-combusted in an oven (450°C) to rid them of potential contaminants. Reserved water samples (10L carboys) were filtered for chlorophyll *a* (Chl *a*)

(not analysed), SPM, particulate organic matter (POM) for SIA, particulate TotHg (PTotHg) and particulate MeHg (PMeHg), using a handheld Teflon filtration system. Quartz fibre filters (Whatman QMA; 2.2µm) were used for PTotHg and PMeHg, while glass fibre filters (Whatman GF/F; 0.7µm) were used for Chl *a*, SPM and POM. The GF/F filtrate was then further processed using a 0.2µm polycarbonate filter for analysis of dissolved nutrients, DOC and DOM characterisation. All filters were then wrapped with aluminium foil, stored in a PE bag and frozen at -20°C. The volume of water filtered varied depending on the sample (Appendix C).

2.3.2 Fish dissections

Fish were defrosted at the NTNU Department of Biology for approximately five hours prior to dissection. This was done to achieve a semi-thawed state, in order to make sample processing easier. The mass (g), length (cm), sex and life stage (adult vs. juvenile) was determined for each fish prior to conducting stomach content analysis (Appendix D). Values for length are reported as standard length, as this method can be applied to both juvenile and adult fish (Kahn et al, 2004). Stomach content analysis was carried out by making an incision across the underbelly of the fish with a sterile stainless-steel scalpel. The stomach was then removed and inspected for any prey material, which identification when possible. Livers (not analysed) and dorsolateral muscle tissue samples were collected, and muscle tissue was divided into subsamples for SIA and Hg analysis (TotHg). Tissue samples were wrapped in aluminium foil, stored in PE bags and kept frozen at -20ºC.

2.3.3 Macroalgae processing

Subsamples of approximately 5cm^2 were taken from the frond of whole macroalgae samples which had previously been rinsed with deionised water and frozen at -20°C. For some metals, variations in concentration are known to occur depending on which part of the macroalgae is sampled. However, this variation has not been shown to influence the concentration of Hg, therefore frond subsamples are considered as representative of the full plant (Burger et al, 2007).

2.3.4 Freeze-drying

Select filters (POM, PMeHg and PTotHg) plus all biotic and sediment samples were batch freeze dried using a Leybold-Heraeus GT2 freeze dryer at the NTNU Department of Biology. All samples were left to dry for three days. Once dry, all samples were transferred to a silica gel desiccator until further processing.

2.3.5 Homogenisation

Freeze-dried biotic samples for SIA and Hg analysis were homogenised using an agate mortar and pestle. This technique is favoured as it prevents contamination of samples used for metal analysis (Thompson & Bankston, 1970). In between each sample, equipment was washed with deionised water and then dried using microscope lens tissue. For freeze-dried samples which had a paper like texture (e.g. holothurians), clean stainless-steel scissors were used to first divide the sample into smaller fractions before using the pestle and mortar.

2.3.6 Acidification

Homogenised subsamples for SIA which had a high carbonate content (e.g. shelled fauna, sediments) were acidified using 1M HCl since carbonates in unacidified samples can lead to inaccurate $\delta^{13}C$ values (Harris et al, 2001). Due to alternating levels of carbonate for each sample, the volume of acid needed to complete this step was variable. Once there was no visible bubbling of the sample after addition of the acid, the process was complete. These samples were then freeze-dried for one day.

2.4. Laboratory analysis

2.4.1 Water chemistry

Analysis of surface water samples for various analytes (ass detailed in section 2.2.2) was conducted at the Norwegian Institute for Water Research (NIVA) using accredited and standardised methods (Skarbøvik et al, 2016). Determination of $SUVA_{254}$ was based on U.S. EPA method 415.3 (U.S. EPA, 2009)

2.4.2 Stable isotopes

Sample preparation for stable isotope analysis (weighing with a microbalance and packing of samples into tin (Sn) capsules) was conducted at The University of Tromsø. Approximately 1mg of material was weighed out for biological samples, while 10-15mg was used for sediments. If little material was present for biological samples, 0.25mg was used instead. For every 10^{th} sample, a duplicate was included. For filters, each sample was packed whole using 8x10mm Sn capsules. Samples which had high carbonate concentrations were sent in two batches, one acidified subsample and one unacidified subsample. This was done to provide a value for unacidified $\delta^{15}N$ and a value for acidified $\delta^{13}C$ (as discussed in section 2.3.6). All packaged samples were then shipped to the University of California Davis Stable Isotope Facility where they were analysed for $\delta^{13}C$ and $\delta^{15}N$ using an Elemental Analyser/Isotope Ratio Mass Spectrometer. Replicates were included every 10^{th} sample and expression of

stable carbon and nitrogen isotopes was expressed using international standards for carbon (Vienna PeeDee Belemnite) and nitrogen (atmospheric N).

2.4.3 Total Mercury

Determination of TotHg in surface water samples was done at NIVA through oxidation, purge and trap and cold vapour atomic fluorescence spectrometry (CVFAS) in accordance to EPA method 1631 (U.S. EPA, 2002). Measurement of TotHg in filters, biotic and sediment samples was conducted using a Direct Mercury Analyser (DMA-80; Milestone, Shelton, Conneticut) at Akvaplan-Niva, Tromsø. Samples which were analysed for TotHg included macroalgae subsamples, amphipods, abundant benthic fauna, fish dorsolateral tissue, sediments and filters. This instrument operates in accordance to EPA method 7473 (U.S. EPA, 1998). The procedure involves thermal decomposition and associated amalgamation of Hg through use of a gold trap, then sequential purging of evaded Hg which can be detected via atomic absorption (U.S. EPA, 1998). The sample mass used for analysis was 0.04-0.05 g for biological material and 0.2 g for sediments, while filters were analysed whole. All samples were measured in pre-cleaned sample boats, which had been combusted using the instruments integrated cleaning programme.

Quality assurance and quality control was tested by the certified reference materials (CRMs) DORM-4 (dogfish muscle tissue; n=3) and MESS-3 (sediment samples; n=3). Recoveries for DORM-4 and MESS-3 were shown to be within the given range for TotHg with 96% and 91% recovery respectively. Blanks (n=2; mean TotHg concentration of 0.009 ng), blank boats (n=2; mean TotHg concentration of 0.006 ng) and blank filters (n=2; mean TotHg concentration of 0.05 ng) were also included in the analysis to control for any potential carry over effect between samples, or background contamination of sample boats and filters.

2.4.4 Methylmercury

Analysis of MeHg was conducted on abundant lagoon benthic samples at the Environmental Science and Analytical Chemistry Department at Stockholm University using a Methyl Mercury Analyser (2700 Methyl Mercury Auto-Analysis System, Tekran, Canada). Procedures were based on slightly altered methodology presented in Hintelmann & Nguyen (2005) and Braaten et al (2014). The sample mass used for analysis was ~0.03 g of biological material. The instrument operates by purging samples with nitrogen gas and trapping volatile Hg onto a Pyrex glass tube via absorption. Trapped volatile Hg is then transported using and internal carrier gas and detected using CVAFS, were free Hg atoms are ‘excited’ by exposure

to ultraviolet light. The amount of radiation produced at a specific wavelength is proportional to the MeHg contained within the sample. All procedures were done in accordance to EPA method 1630 (U.S. EPA, 1998).

Quality assurance and quality control measures for MeHg analysis included the use of method blanks (*n*=3; mean MeHg concentration of 0.2ng/L), the CRM TORT-2 (lobster hepatopancreas; *n*=2), which was within 1% of the certified value for MeHg (0.137 ± 0.012 mg/kg), matrix spikes (*n*=2; with 84% and 89% recovery respectively) and the inclusion of replicate samples (*n*=3; relative percent difference ranged from 1.3-19.6%).

2.5. Data analysis

All statistical analysis was performed in R studio (version 3.4.2) using the *car* package (Fox et al, 2007), while all graphics were produced with *ggplot2* (Wickham, 2011; R Development Core Team, 2017).

Univariate approaches were applied to the data set. Prior to statistical analysis, all data were log transformed (Log_{10}) to approximate a normal distribution. Data were then checked for normality and homoscedasticity using Shapiro-Wilk and Levene's tests, with a level of significance set to p=0.05. Due to the unbalanced nature of the data set, assumptions of normality were violated, therefore non-parametric statistical tests were applied to the data.

In order to test if there were significant differences between the TotHg concentrations of lagoon taxa (plus POM), a Kruskall-Wallis test was used. A post-hoc Dunn's test with Bonferroni adjustment was then used to identify which groups were significantly different from each other. In order to explore if there were significant differences in Hg accumulation of sculpin, a Mann-Whitney U test was applied to explore the relationship of sex and the relationship of life stage. Linear regression was applied to test the relationship between TotHg and $\delta^{15}N$ for the whole lagoon food web, plus to test the TotHg concentrations in sculpin with various biometric parameters (length, weight, condition factor). Normality of the residuals was also tested through use of the Shapiro-Wilk test. The dependant variable for all linear regression models was TotHg concentration.

2.6. Trophic level and biomagnification calculations

Trophic level (TL) of lagoon fauna was calculated in relation to the TL of benthic bivalves ($TL_{bivalve}$). This group was assumed to be the primary consumers of this system based on the taxa that were sampled and their feeding strategy (filter feeders). Bivalves were therefore

assigned a TL of 2.0. The TL of other lagoon fauna were calculated using a modified version of the equation presented in Fisk et al. (2001):

$$i)\ TL_{consumer} = TL_{bivalve} + \frac{(\delta^{15}N_{consumer} - \delta^{15}N_{bivalve})}{3.4}$$

Where $TL_{consumer}$ and $\delta^{15}N_{consumer}$ respectively represent the trophic level and stable nitrogen isotope value of lagoon fauna in relation to benthic bivalves. The value of $\delta^{15}N_{bivalve}$ represents the mean stable nitrogen isotope value of all sampled benthic bivalves, which equates to 7.0‰. The average enrichment of $\delta^{15}N$ ($\Delta^{15}N$) per TL was assumed to be 3.4‰, which is based on an assigned value for the Barents Sea region (Søreide et al, 2006).

Trophic level normalised biomagnification factors (BMF_{TL}) were calculated for TotHg and MeHg. These are expressed as the contaminant concentration (in this case Hg as TotHg or MeHg) between predator and prey species in association with TL, as detailed in Fisk et al. (2001):

$$ii)\ \mathrm{BMF_{TL}} = \frac{(\mathrm{Hg_{predator}}/\mathrm{Hg_{prey}})}{(\delta^{15}N_{predator}/\delta^{15}N_{prey})}$$

Here, $Hg_{predator}$ and Hg_{prey} represent the Hg (TotHg or MeHg) concentration in ng/g dw of consumer and prey fauna, while $\delta^{15}N_{predator}$ and $\delta^{15}N_{prey}$ are the mean $\delta^{15}N$ values for predatory fauna and prey material respectively. BMF's with a value >1 indicate that Hg has biomagnified in predatory species following prey consumption. When BMFs have a value <1, no biomagnification is shown to occur between predator and prey species.

In addition, the trophic magnification slope (TMS) was determined for the entire lagoon food web. This is based on the linear regression of logarithmically transformed contaminant data and mean $\delta^{15}N$ values for all organisms represented in the lagoon food web:

$$iii)\ Log_{10}(TotHg) = a + b(\delta^{15}N)$$

The dependent variable is the logarithmically transformed Hg data, which is represented as $Log_{10}(TotHg)$ in the equation, while the explanatory variable is $\delta^{15}N$ - the mean $\delta^{15}N$ value for each component of the lagoon food web. The TMS equates to the slope (*b*) which is produced as a result of the regression between $Log_{10}(TotHg)$ and $\delta^{15}N$, with the y-intercept being represented as *a*.

Finally, a trophic magnification factor (TMF) was calculated for the lagoon food web. The calculated TMF represents the rate of Hg biomagnification throughout the entirety of the food

web. This is calculated using the slope (b) of the linear regression between $Log_{10}(TotHg)$ and TL (as derived from Equation i):

$$iv)\ TMF = 10^{b}$$

3. Results

3.1. Physical and chemical parameters

3.1.1 Hydrography

Within Richardlaguna, there are two distinct hydrographic settings. These are represented through the physical characteristics of the five sampling stations as seen in Table 3.1. The nearshore stream inlets are distinctly freshwater, having salinity values <0.5 (ppt). Salinity values for the other stations had a marine signal with salinity values >30 (ppt). Both the temperature and pH of all stations show little variation. Variability was shown for turbidity with a range of 2.0-10.1 (NTU). Turbidity was highest in the nearshore stream inlets. Data from the CTD profiles for the lagoon basin and outlet indicate that there is a very thin freshwater layer present at the surface (~0.5m) of these stations (Appendix E). In both cases this is subsequently followed by more saline water (>30 ppt) for the remainder of the water column.

Table 3.1. Select physical parameters of lagoon sampling sites.

Sample I.D.	Location	Temperature (^{o}C)	Salinity (ppt)	pH	Turbidity (NTU)
In1	Stream Inlet 1	7.1	0.1	8.1	6.7
In2	Stream Inlet 2	6.1	0.4	8.0	10.1
Cen	Basin	5.9	30.8	7.8	3.1
Out	Outlet	7.9	33.8	7.9	2.0
Mar	Outer Lagoon	7.9	35.0	7.9	2.2

Note: Physical parameters were derived from surface water samples, which were measured on the field with a handheld multi-sensor. Turbidity values are based on the average of three measurements.

3.1.2 Water chemistry

Nutrient concentrations varied between sample locations (Table 3.2). Concentrations of TN, NO^{2-}/NO^{3-} and SiO_4 were highest at the nearshore stream inlets. The highest concentration of NO^{2-}/NO^{3-} was 134.01 µg/L and was observed at station In1. This value was two orders of magnitude higher than the lowest observed NO^{2-}/NO^{3-} concentration, which was 3.92 µg/L at the marine station. Station In2 had a SiO_4 concentration of 2076 µg/L. This value was almost three times as higher than the concentration observed at In1 inlet and eighteen times higher than the value for the marine station. Both TP and PO_4^{3-} concentrations were highest at the marine station and lowest at the stream inlets. Concentrations of NH_4^+ varied across all stations and was highest at station In1 with a concentration of 16.73 µg/L, while the lagoon central basin had the lowest concentrations at 6.08 µg/L.

Table 3.2. Select water chemistry parameters measured in surface water samples from Richardlaguna.

I.D.	Location	TN (µg/L)	NH_4^+ (µg/L)	NO^{2-} /NO^{3-} (µg/L)	TP (µg/L)	PO_4^{3-} (µg/L)	SiO_4 (µg/L)	SPM	PTotHg (ng/g)	Aqueous TotHg (ng/L)
In1	Stream Inlet	280	16.73	134.01	3.49	1.21	700	6.3	5.09	2.63
In2	Stream Inlet	200	8.49	120.74	3.61	1.27	2076	24.4	7.61	2.11
Cen	Basin	130	6.08	55.59	20.4	3.79	184	29.2	0.90	0.42
Out	Outlet	140	7.40	5.52	18.5	5.27	135	31.1	1.42	0.31
Mar	Outer Lagoon	120	12.44	3.92	16.95	6.32	111	29.7	0.68	0.37

High SPM values were observed for all stations except for In1 (Figure 3.1a). The specific ultraviolet absorbance at 254nm ($SUVA_{254}$) varied across study locations (Figure 3.1.b). Higher $SUVA_{254}$ values were observed for the nearshore stream inlets and the lagoon outlet, while lower $SUVA_{254}$ values were observed for the lagoon basin and marine station. DOC typically made up approximately one third of the TOC present for all sampling stations (Figure 3.1.c), with the highest TOC concentration observed at station In1. Values for aqueous TotHg (Figure 3.1.d) were higher at the nearshore stream inlets (>2.0 ng/L) and lower for all other stations (<0.5 ng/L). Concentrations of C (Figure 3.1e) and Hg (Figure 3.1f) in the particulate phase (in ng/g of SPM) were highest at the nearshore stream inlets and lowest at the outer marine station.

3.1.3 Sediment characteristics

The mean temperature, pH and redox potential of lagoon surface sediments (collected from the lagoon central basin) was 10.3 (°C), 6.8 and -14.2 (mV) respectively. The TotHg concentration for lagoon sediments had a mean concentration of 16.5 ng/g dw.

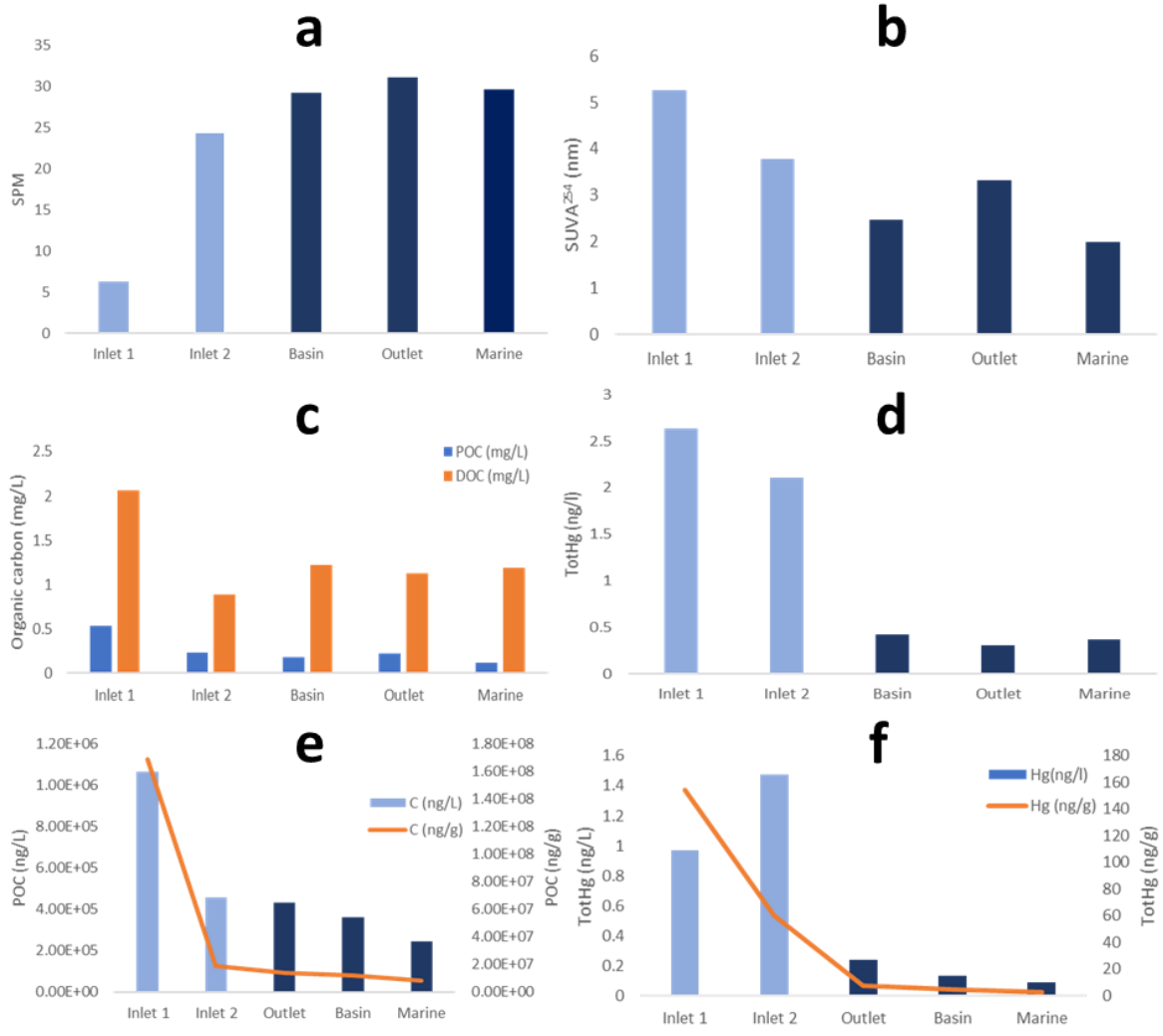

Figure 3.1. Bar plots displaying surface water data for a) suspended particulate matter b) specific ultraviolet absorbance at 254nm c) concentration of particulate and dissolved organic carbon (mg/L) d) concentration of total mercury (ng/L) e) particulate organic carbon in the particulate phase and f) total mercury in the particulate phase for each lagoon station.

3.2 Lagoon food web structure

Five species of macroalgae were identified, which were *Alaria esculenta* (*n*=1), *Laminaria sp.* (*n*=1), *Desmarestia sp.* (*n*=1), *Fucus sp.* (*n*=1) and *Saccharina latissima* (*n*=1). Amphipods were solely composed of *Gammarus sp.* (*n*=384). The benthic infaunal community was largely represented by polychaete worms and bivalves, with *Spionidae sp.* (*n*=265), *Brada villosa* (*n*=119) and *Thyasira sp.* (*n*=67) representing the three most abundant organisms. In addition, four other polychaete worms (*Maldanidae sp.*, *Terebellidae sp.*, *Polynoidea sp.* and *Scoloplos armiger*) and four other bivalves (*Hiatella arctica*, *Liocyma sp.*, *Mya sp.* and *Macoma calcarea*) were also collected. Lagoon benthic infaunal community structure was further composed of two gastropod species (*Cylichna sp.* and *Unknown gastropod*), one holothurian species (*Chiridota laevis*) and one species of priapulid worm

(*Priapulus caudatus*). Fish caught included two species of sculpin - shorthorn sculpin (*Myoxocephalus scorpius, n*=17) and Arctic staghorn sculpin (*Gymnocanthus tricuspis, n*=1).

3.3. Stable isotopes and trophic structure

3.3.1 Isotopic values

All biotic samples (plus POM) were analysed for SIA. Sculpin had the highest overall $\delta^{15}N$ values of all sampled lagoon fauna, with a range of 12.4‰-14.1‰ (Figure 3.2). Of the benthic infauna community, the priapulid worm *Priapulus caudatus* had the highest $\delta^{15}N$ value (11.6‰). This was followed by a small cluster, composed of both gastropods (*Cylichna sp.* and *Unknown gastropod*) and one polychaete (*Polynoidea sp.*), which had $\delta^{15}N$ values of 10.1‰, 10.6‰ and 10.5‰ respectively. Almost all the other benthic infauna (except for three bivalves), with the addition of *Gammarus sp.*, tended to form another distinct cluster with a $\delta^{15}N$ range of 7.4-9.4‰. A final smaller cluster for the remaining three bivalves is shown, which have a $\delta^{15}N$ range of 6.4-6.9‰. POM had the highest variability in $\delta^{15}N$ with a range of 2.5-5.8‰ across the study sites, while macroalgae values were consistent except for *Laminaria sp.* (4.8‰).

For $\delta^{13}C$, values for lagoon biota ranged -21.7‰ in *Liocyma sp.* to -18.0‰ in Arctic staghorn sculpin. The mean $\delta^{13}C$ value for shorthorn sculpin was -18.4‰, while the value for the single Arctic staghorn sculpin was -18.0‰. For benthic infauna, the largest variation in $\delta^{13}C$ is seen for bivalves, which has a range of -21.6‰ to -18.0‰. The highest variability in $\delta^{13}C$ is shown for macroalgae, with values ranging from -23.5‰ for *Desmarestia sp.* to -16.4‰ for *S. latissima*.

3.3.2 Trophic level

Due to their status as primary producers, all sampled macroalgae were assigned a trophic level (TL) of 1. Similarly, all bivalves were given a TL equating to 2 as they were assumed to be primary consumers (Dunton et al, 2012). This group of organisms had the lowest overall $\delta^{15}N$ values compared to all other sampled fauna (Figure 3.2.), therefore reinforcing this assumption. Overall the TL derived for lagoon fauna ranged from 1-4.2 (Table 3.3), with macroalgae representing the lowest level and sculpin represented the highest level.

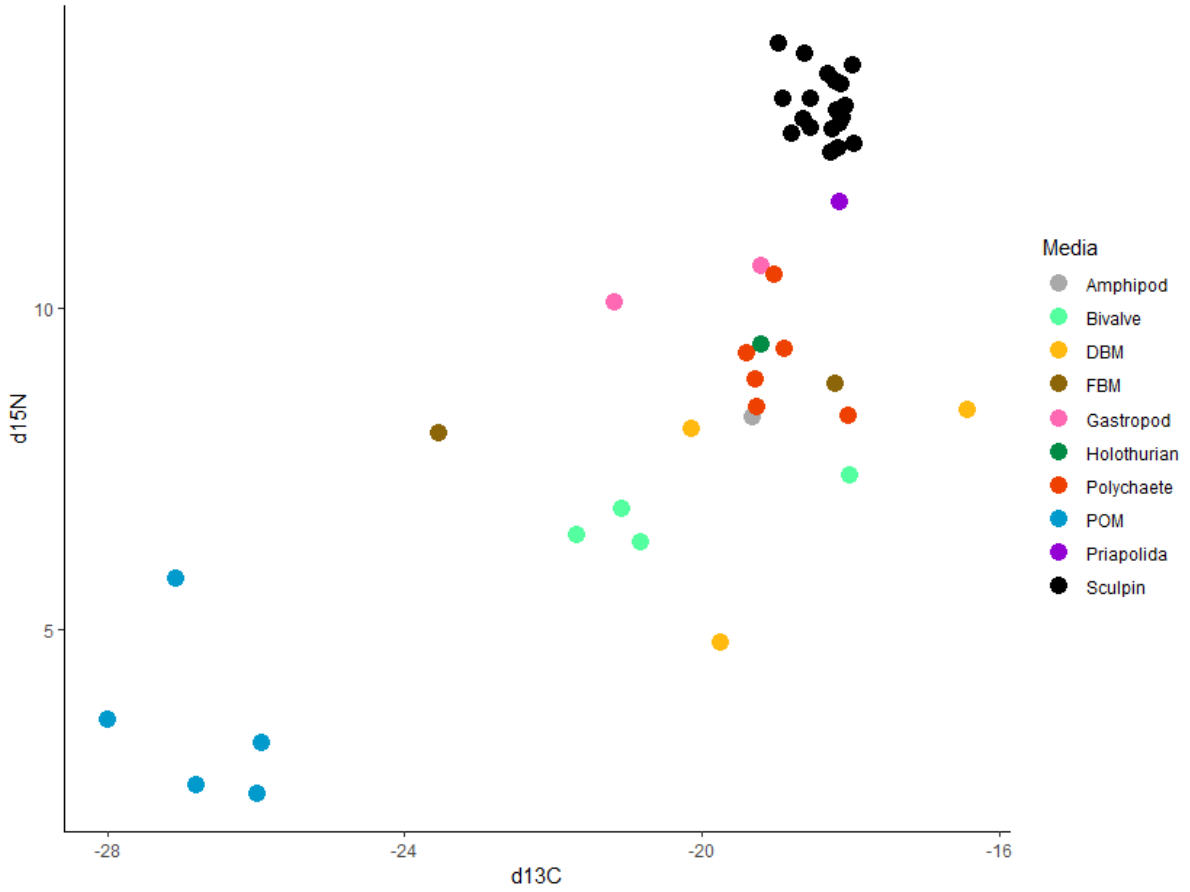

Figure 3.2. Stable isotope biplot showing values of $\delta^{13}C$ (d13C; x-axis) and $\delta^{15}N$ (d15N; y-axis) for particulate organic matter and all sampled lagoon fauna. Due to suspected incomplete acidification, the bivalve *Thyasira sp.* was omitted from this plot. Different media (i.e. POM, flora and fauna) are separated into appropriate categories which are represented by colour.

3.4. Concentrations of Hg in lagoon flora and fauna

The mean concentrations of TotHg (ng/g dw) and MeHg (ng/g dw) are summarised for select taxa in Table 3.3. There were statistically significant differences in TotHg concentrations between lagoon taxa (plus POM) (Kruskall-Wallis test; $p<0.0001$) (Figure 3.3). Further exploration revealed that there were significant differences between POM and sculpin (Dunn's test; $p=0.001$) and macroalgae and sculpin (Dunn's test; $p=0.003$). All other interactions were not statistically significant (Appendix F).

3.3.1 Macroalgae

TotHg concentration in macroalgae were low in both groups with drift brown macroalgae (DBM) having a range of 5-12 ng/g dw and fresh brown macroalgae (FBM) having a range of 6-8 ng/g dw. *Laminaria sp.* had the highest TotHg concentration (12 ng/g dw), while *A. esculenta* (5 ng/g dw) had the lowest.

3.3.2 Amphipoda

The TotHg concentration in the pooled *Gammarus sp.* sample was 43 ng/g dw, while the value for MeHg was 16 ng/g dw. The concertation's of TotHg and MeHg were established from taking the average of three replicate samples for TotHg and two replicate samples for MeHg.

3.3.3 Abundant benthic fauna

The concentration of TotHg in abundant benthic fauna ranged from 17-80 ng/g dw, with the lowest value being for the priapulid worm *P.caudatus* and the highest value being for the polychaete *Terbellidae sp.* The remaining benthic fauna had TotHg concentrations <40 ng/g dw, except for the polychaete *Maldanidae sp.* and the bivalve *H.arctica* which had concentrations of 60 ng/g dw and 49 ng/g dw respectively. Concentrations of MeHg were generally <10 ng/g dw, except for *H.arctica*, *Polynoidea sp.* and *Maldanidae sp.* The lowest MeHg concentration was observed for the holothurian *C.laevis* (2 ng/g dw), while the highest concentration was found for *H.arctica* (25 ng/g dw). The percentage of Hg present as MeHg (%MeHg) was below 50% for all benthic fauna except for *H.arctica* (Appendix B).

3.3.4 Sculpin

Of all taxa sampled, sculpin had the highest TotHg concentrations (168±84 ng/g dw, n=18). TotHg concentrations in shorthorn sculpin were variable and ranged from 69-418 ng/g dw, while the single Arctic staghorn sculpin had a TotHg concentration of 120 ng/g dw. Positive relationships between TotHg and length (R^2=0.51, p=0.001) as well as TotHg and weight (R^2=0.33, p=0.01) were observed, while a negative relationship was found between condition factor (k) and TotHg (R^2=0.35, p=0.01) (Figure 3.4). No significant differences were observed for the effects of sculpin sex (Mann-Whitney U test; p=0.5) or life stage (Mann Whitney U test; p=0.4) on TotHg concentrations (Figure 3.5).

Table 3.3. Mean TotHg and MeHg concentrations (ng/g dw), percentage of TotHg present as MeHg (%MeHg), stable isotopes, calculated trophic level and ecological information on select taxa from Richardlaguna. DBM stand for drift brown macroalgae and FBM stands for fresh brown macroalgae.

Organism	Tissue Analysed	*n*	Feeding Strategy	Mean ± SD TotHg (Range)	Mean MeHg	%MeHg	Mean ± SD $\delta^{13}C$	Mean ± SD $\delta^{15}N$	TL
Macroalgae									
DBM	Frond/Blade	3	Autotroph	10 ± 4 (5-12)	N/A	N/A	-18.9 ± 2	9.0 ± 3	1.0
FBM	Frond/Blade	2	Autotroph	7 ± 2 (6-8)	N/A	N/A	-20.9 ± 4	9.9 ± 1	1.0
Bivalvia									
Hiatella arctica	Soft tissue	*1	Filter/suspension	49	25	52	-21.5	6.9	2.0
Polychaeta									
Spionidae sp.	Whole body	*1	Surface deposit	28	3	11	-18.9	9.4	2.8
Polynoidea sp.	Whole body	*1	Scavenger/predator	29	12	43	-19.0	10.5	3.1
Terbellidae sp.	Whole body	*1	Surface deposit	80	3	4	-19.4	9.3	2.8
Maldanidae sp.	Whole body	*1	Surface deposit	60	16	26	-19.3	8.9	2.6
Brada villosa	Whole body	*1	Surface deposit	39	3	8	-18.0	8.3	2.5
Priapulida									
Priapulus caudatus	Whole body	*1	Scavenger/predator	17	6	37	-19.3	11.6	3.5
Holothuroidea							-19.2	9.4	2.8
Chiridota laevis	Whole body	*1	Scavenger	30	2	6			
Amphipoda									
Gammarus sp.	Whole body	*1	Grazer/scavenger	43	16	42	-19.3	8.3	2.5
Chordata									
Myoxocephalus scorpius	Dorsolateral tissue	17	Predator	170 ± 86 (69-418)	N/A	N/A	-18.2 ± 0.3	13.1 ± 0.5	3.7-4.2
Gymnocanthus tricuspis	Dorsolateral tissue	1	Predator	120	N/A	N/A	-18.0	13.8	4.1

Note: N/A = non-applicable, feeding strategy is based on information from the Arctic Traits database (Degen & Faulwetter, 2019). Pooled samples are indicated by (*), were the number of individuals pooled for each sample is presented in Appendix B.

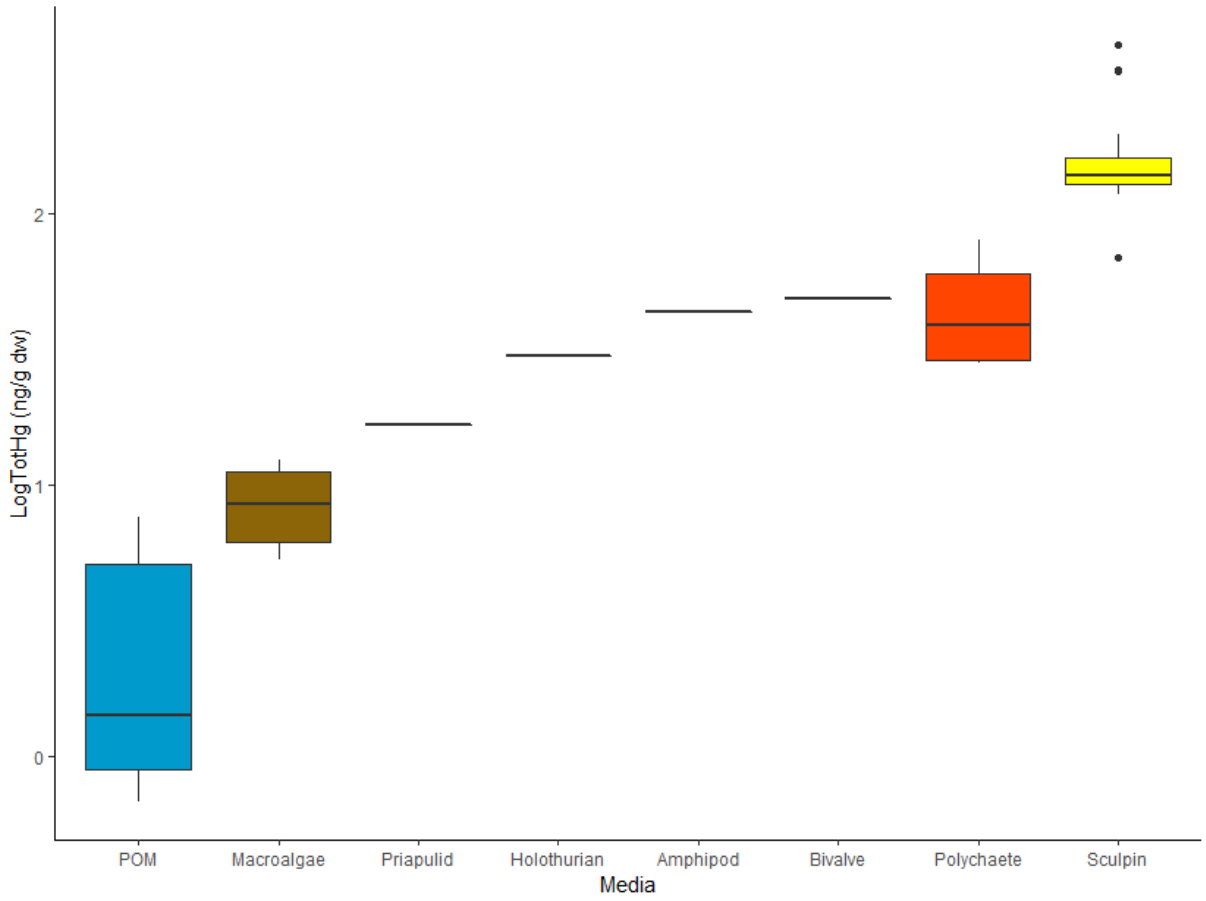

Figure 3.3. Boxplots showing the Log TotHg concentrations (ng/g dw) for select lagoon fauna and POM. The plot shows medians (thick horizontal lines in the boxes), maximum and minimum values within a 1.5 inter quartile range (vertical lines) and outliers (black dots).

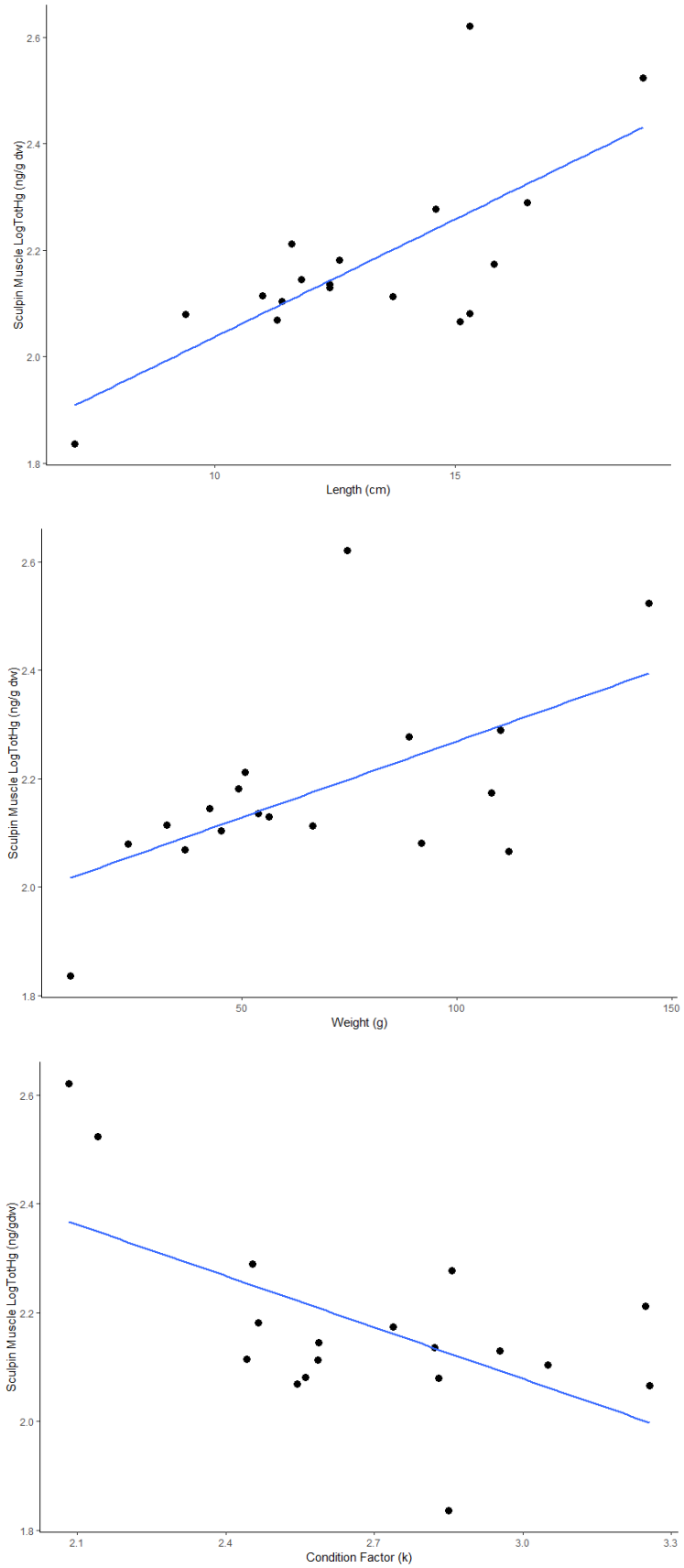

Figure 3.4. Linear regressions focusing on the relationships between Log10 TotHg concentrations (ng/g dw) of sculpin (*n*=18) in association with: a) length (cm) (log-TotHg = 0.044 x Length + 1.594, R^2=0.051, *p*=0.001) b) weight (g) (log-TotHg = 0.003 x Weight + 1.988, R^2=0.33, *p*=0.01) c) condition factor (k) (log-TotHg = -0.315 x K + 3.022, R^2=0.35, *p*=0.01).

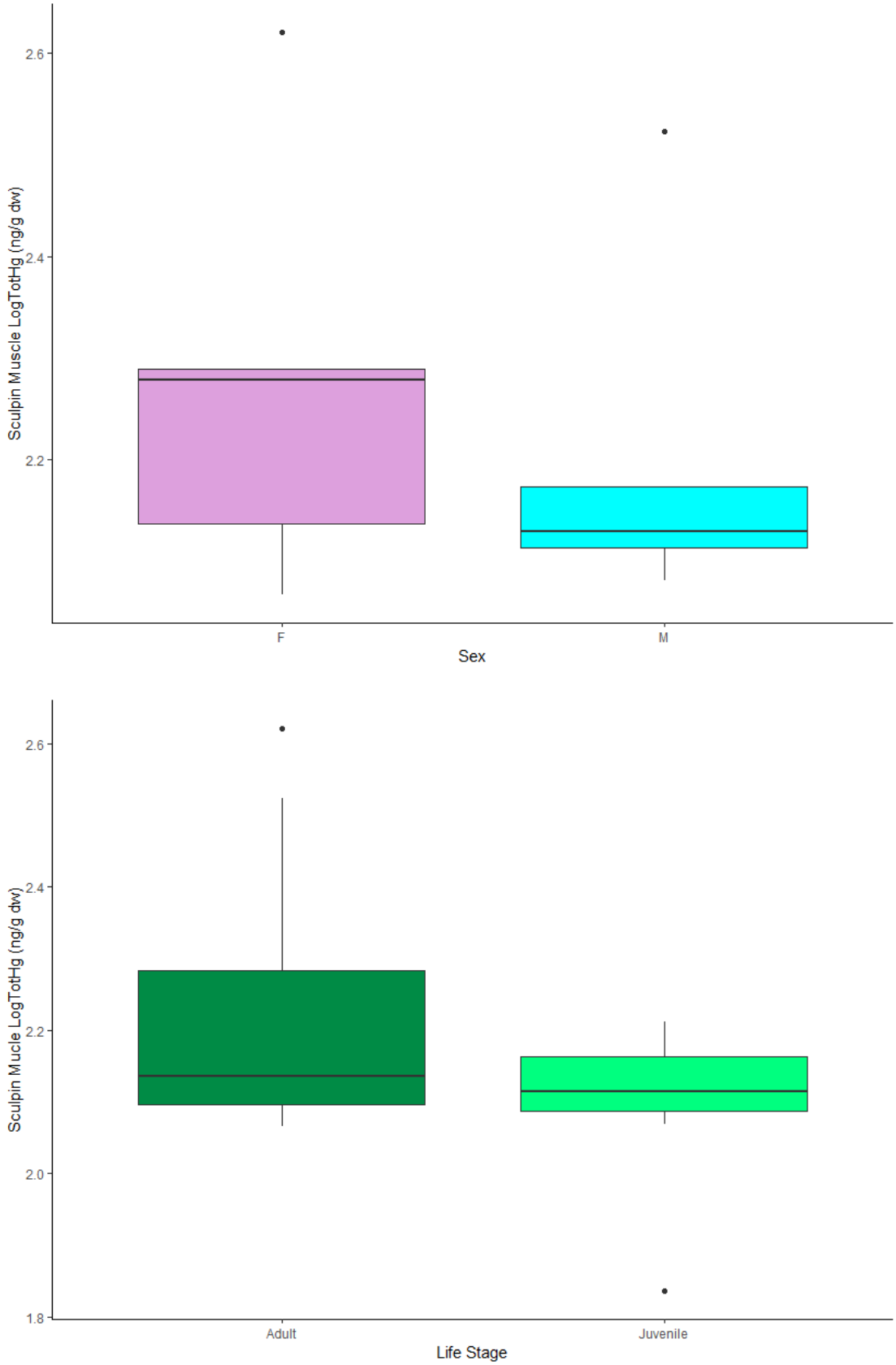

Figure 3.5. Boxplots comparing sculpin Log TotHg concentrations (ng/g dw) with sex (top) and sex (life stage). Sex is separated into males (M; *n*=5) and females (F; n=*6*), while life stage is separated into adults (n=*11*) and juveniles (n=*7*). The plots show medians (thick horizontal lines in the boxes), maximum and minimum values within a 1.5 inter quartile range (vertical lines) and outliers (black dots)

3.5. Biomagnification of Hg

3.5.1 Biomagnification factors

A BMF was calculated for sculpin and their prey (*Gammarus sp.)* as revealed by stomach content analysis. By using the average TotHg concentration (ng/g dw) and $\delta^{15}N$ values for both sculpin and *Gammarus sp.* respectively in the BMF calculation, a value of >1 was achieved (BMF = 2.3). This therefore indicated that TotHg biomagnifies for this specific predator/prey interaction.

3.5.2 Trophic magnification factors

TMS and TMF were calculated based on all the sampled species which were analysed for TotHg. The TMS value (0.18) is derived from the linear regression between Log TotHg (ng/g dw) and $\delta^{15}N$ (Figure 3.6). TMF value was >1 (TMF = 3.4), which indicates that TotHg biomagnifies with each successive trophic link within the lagoon food web, based on the fauna sampled. This was from the linear regression between Log TotHg (ng/g dw) and TL (log-TotHg = 0.5280 x TL +0.1239, R^2 = 0.82, p<0.0001) (Appendix G).

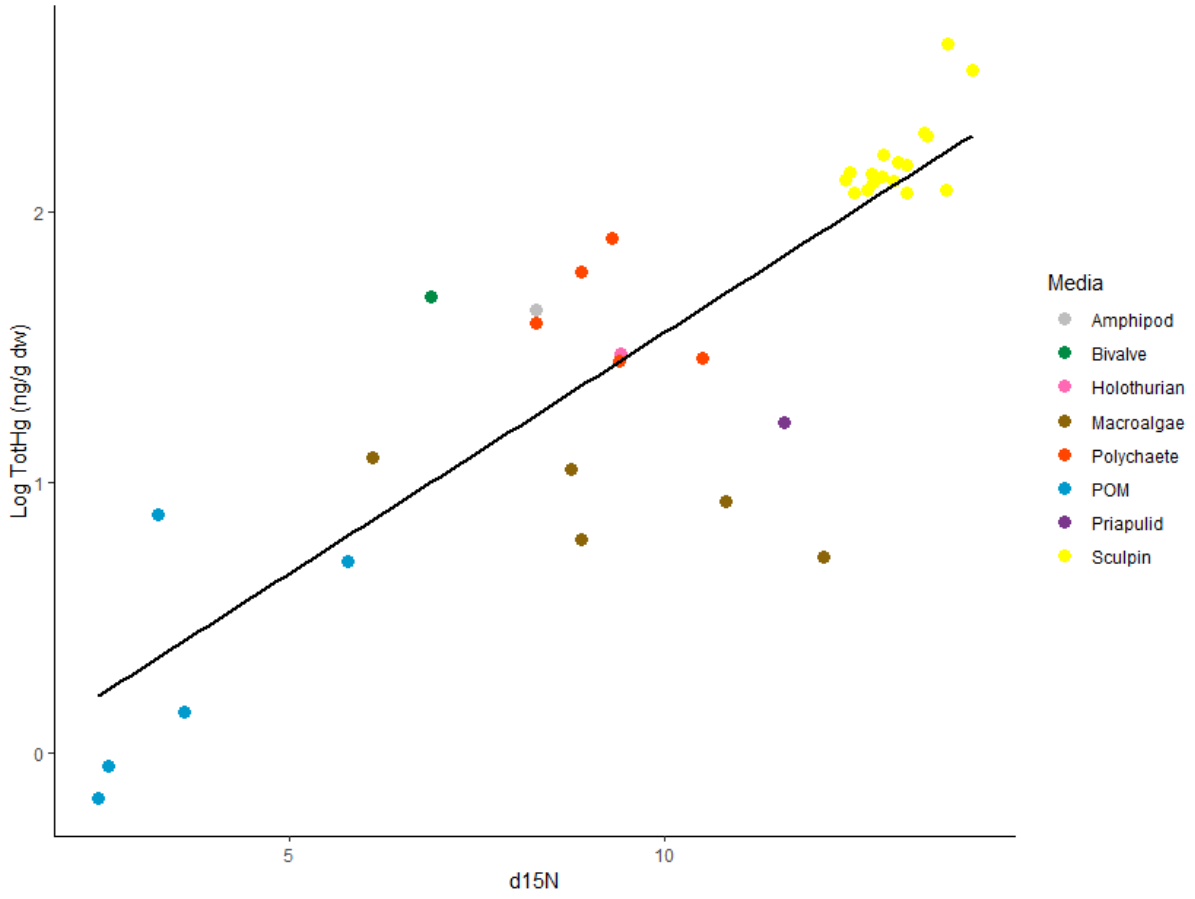

Figure 3.6. Linear regression of the relationship between Log TotHg (ng/g dw) versus $\delta^{15}N$ (%) in POM and select lagoon fauna (log-TotHg = 0.178 x $\delta^{15}N$ – 0.228, R^2=0.73, p<0.0001, *n*=36).

4. Discussion

4.1. Stream inlets are sources of tDOM and Hg

Rivers and streams are significant sources of Hg and organic carbon to Arctic coastal environments (Fisher et al, 2012; Soerensen et al, 2016), and our findings suggest that this is also true for the stream inlets in Richardlaguna. Lagoon inlet streams are important sources of Hg, as aqueous Hg concentrations in inlets were approximately 4-5 times higher compared to the lagoon central basin, lagoon outlet and from outer marine stations. This was also observed for TOC at one of the nearshore stream inlets (In1), where the TOC concentration was approximately twice the value for all other sampling stations. Interestingly, SPM values were lower in the lagoon inlet streams than at the lagoon and marine stations, even though we found enhanced concentrations of both aqueous and particulate Hg, plus TOC. This suggests that particles at the stream inlet stations are more enriched in Hg and TOC in comparison to the other lagoon stations with higher SPM values.

The higher concentrations of particulate and aqueous Hg in the inlet streams is likely linked with the enhanced export of tDOM. The relationship between Hg and OM is well established, in that the transportation of TotHg and MeHg in watersheds is tightly associated with the mobilisation, transport and fate of DOC and particulate organic carbon (POC) (Schuster et al, 2008; Shanley et al, 2008). When Hg^{2+} is present in the environment, it is capable of actively binding with DOM through bonding with reduced sulphur sites (e.g. thiols) present on the OM molecules (Skyllberg et al, 2000; Haitzer et al, 2003). The chemical composition of DOM is therefore an important factor to consider in relation to the movement of Hg from terrestrial to aquatic environments. For example, the rate of Hg^{2+} sorption is greater when organic matter is composed of fulvic and humic acids which are S-rich and can readily form complexes with Hg (Skyllberg et al, 2000).

The lower particulate and aqueous Hg concentrations reported for the outer marine station could be due to restricted movement of particles from the nearshore stream inlets. Lagoons often have lower exchange with the open marine environment, and can have relatively long water residence times, which can create an environment that is conductive to loss of terrestrially derived material through sedimentation. This can be by direct sedimentation of particulate matter (plus particle associated Hg) or through flocculation (movement of OM from the dissolved to particulate phase) once terrestrial material encounters saline water. In aquatic environments, bottom sediments are often the ultimate sink for Hg. It is estimated that

only 6% of Hg exported from Arctic rivers reaches the open ocean (Zhang et al, 2015). This is consistent with the results of the current study, which observed the lowest aqueous and particulate Hg concentrations at the outer marine station. It is likely that most of the Hg being transported to the lagoon from the surrounding catchment eventually ends up stored in lagoon sediments.

The variability in $SUVA_{254}$ found in this study indicates that there are differences in the aromaticity of organic matter between lagoon stations. O'Donnell et al (2016) indicate that higher $SUVA_{254}$ and DOC concentrations in rivers/streams indicates that there are potential influences from ice-rich permafrost (O'Donnell et al, 2016). Our findings showed that values for $SUVA_{254}$ and DOC were highest at the lagoon inlet streams, suggesting that these streams are being influenced by a similar upstream environment (e.g. fine-grained permafrost overlain soils). The differences observed between the two inlet streams may also be attributable to differences in stream catchment characteristics (e.g. topography, land-cover, hydrology, geology).

The data reported in this study likely underestimate the typical concentrations of aqueous and particulate Hg and TOC in this lagoon system (and inlet streams) during the open water season. This is due to the nature of the sampling, which only provides a 'snapshot' view of physiochemical conditions in this system. For example, at the time of sampling (early September), the lagoon was highly saline with only a slight freshwater surface layer. However, we would anticipate that given the restricted exchange with the open marine environment, Richardlaguna is likely to have quite low salinity throughout the whole lagoon area at times of the year with higher terrestrial runoff (e.g. snowmelt, peak glacial melt, large rainfall events), as has been seen in other Arctic lagoon ecosystems (Harris et al, 2017). Similarly, it is expected that there would be a greater influx of Hg and TOC to Richardlaguna in the spring and summer months through enhanced seasonal melting, as is the case for other Arctic coastal systems (Leitch et al, 2007). There may also be a seasonal increase in Hg to the lagoon environment through the presence of migratory fauna (e.g. walrus and seabirds), which could potentially contribute Hg in the form of faeces and guano. Hg in the lagoon benthic environment could also potentially be influenced by the seasonal phytoplankton bloom, which may act as a source of Hg to the benthos once the phytoplankton bloom dies off and settles onto bottom sediments. In order to characterise the influence of seasonality on Hg dynamics in the lagoon environment, future sampling in this area should be conducted throughout the year.

4.2. Lagoon food web structure

Due to lagoon ecosystems being highly influenced by terrestrial runoff, we expected that fauna in Richardlaguna would rely quite strongly on terrestrial carbon sources compared to marine carbon sources, as this has been reported in Alaskan coastal lagoons (Dunton et al, 2012; Harris et al, 2018). However, our $\delta^{13}C$ data contradicted this assumption as we found that lagoon fauna relied heavily on marine carbon sources, though some taxa (polychaetes, bivalves) did show some degree of reliance on terrestrial food sources.

The $\delta^{13}C$ values reported for lagoon fauna are reminiscent of $\delta^{13}C$ values for marine phytoplankton data from Svalbard. These values reported in the present study are also similar to $\delta^{13}C$ values reported during the 2018 seasonal bloom from several stations in Isfjorden on Svalbard (Poste, *unpublished data*). This suggests that at the time of sampling, lagoon faunal stable isotope values still reflect the mDOM from the seasonal bloom or that there are other important marine food sources in the lagoon e.g. macroalgae, microphytobenthos (MPB). Most Arctic marine food web studies often overlook the role of MPB as a source of primary production even though it is prevalent in shallow, coastal regions (Glud et al, 2009). This is primarily due to difficulties in determining MPB $\delta^{13}C$ values as it is difficult to separate MPB from sediment (Oxtoby et al, 2016). However, there is evidence to suggest that $\delta^{13}C$ values for MPB overlap pelagic food sources (e.g. phytoplankton) (Oxtoby et al, 2016), therefore it is possible that fauna in Richardlaguna are selectively relying on MPB as a food source.

Due to the 'snapshot' nature of our sampling, which took place during a low period of terrestrial inputs, there may also be an underestimate regarding the dietary reliance of terrestrial carbon in lagoon fauna with faster tissue turnover times. It is possible that lagoon fauna rely quite strongly on terrestrial derived carbon during times of the year when terrestrial inputs to the lagoon are highest during seasonal melting (June-July), which may not be reflected in our sampling as this occurred later in the season.

The $\delta^{15}N$ values and derived TL's for lagoon fauna for the present study reflect the known feeding strategies of the sampled organisms (Degen & Faulwetter, 2019). This is expressed clearly in lagoon benthic fauna, in that organisms which are active predators or scavengers (e.g. *Polynoidea sp.*, *Priapulus caudatus*) have higher $\delta^{15}N$ values and inhabit higher TL's compared to species which rely on surface deposit and suspension feeding. Amphipods from Richardlaguna had a lower $\delta^{15}N$ value (and estimated TL) compared to other scavenging fauna. This may be due to further layers of complexity in amphipod feeding ecology. For

example, it has been reported that Arctic amphipods can employ various feeding strategies (e.g. active predation/scavenging, deposit-feeding/active predation, phyto-detritivores) (Legeżyńska et al, 2012). Seasonal variation in $\delta^{13}C$ values and $\delta^{15}N$ values has also been reported in amphipods and has been linked to habitat, ontogenetic and seasonal changes (Legeżyńska et al, 2012; Legeżyńska et al, 2014; Skogsberg, *unpublished data*). The $\delta^{15}N$ values of *Gammarus sp.* reported for this study are in line with the values reported by Skogsberg (*unpublished data*) for amphipods (*Gammarus setosus*) collected Adventfjord (Svalbard) in August 2018.

The variation in $\delta^{15}N$ values and TL of sculpin suggest that sculpin inhabiting the lagoon have variable diets, though results from stomach content analysis revealed that diet was solely composed of amphipods (plus one small fish and unidentified well digested material). However, gut content analysis only provides information on recently consumed prey, so does not provide a full representation of an organism's diet. Sculpin are known to predate on prey from higher trophic levels once they reach certain lengths and can be cannibalistic as adults (Ruzycki & Wurtsbaugh, 1999; Laundry et al, 2018). Size-dependant dietary changes have been reported in a study from the Baltic Sea were sculpin <24cm fed on the crustacean *Mysis mixta*, while sculpin >26cm had a diet consisting of isopods (*Mesidotea entomon*) and herring (*Clupea harengus*) (Cardinale, 2000). In the present study we found that that amphipods (*Gammarus sp.*) were the most important component of sculpin diets (at the time of sampling) and is in accordance with other studies (e.g. Moore & Moore, 1974; Lydersen et al, 1989; Dick et al, 2009). All the sculpin sampled in this study were <26cm, so it is possible that size-dependant dietary changes have not yet occurred for these individual fish. However, sculpin are fairly opportunistic feeders and diet depends strongly on prey availability, therefore drawing comparisons from other dietary studies is difficult.

4.3. Hg concentrations in lagoon biota are comparable with other Arctic coastal environments

The concentrations of TotHg in lagoon biota were found to be in accordance with other studies focusing on Arctic coastal environments (Table 4.1.). Generally, the concentrations found in this study were similar or higher than the values reported from other studies from the Svalbard area. However, concentrations of Hg in lagoon fauna were lower than values reported for fauna from other Arctic regions.

Table 4.1. Comparison of TotHg and MeHg concentrations (ng/g dw) for select fauna sampled in Richardlaguna and other Arctic marine environments from previous studies.

Phylum	TotHg (Lagoon)	TotHg (Other)	MeHg (Lagoon	MeHg (Other)	Location	Reference
Macroalgae						
S. latissima	11	14-40	N/A	N/A	Grønfjorden, Svalbard	Lebedeva et al, 2018
		93±19		N/A	Canadian Arctic	Clayden et al, 2015
Fucus sp.	8	7-27	N/A	N/A	Canadian Arctic	van der Velden et al, 2013
Amphipoda						
Gammarus sp.	43	16-30	16	1-22	Adventfjorden, Svalbard	Skogsberg, *unpublished data*
		232±58		27±9	Canadian Arctic	Clayden et al, 2015
		16-49		N/A	Canadian Arctic	van der Velden et al, 2013
		N/A		2-7	Isfjorden, Svalbard	Poste et al, *unpublished data*
		N/A		0.4-6.2	Alkhornet, Svalbard	Finne*, unpublished data*
Priapulida						
P. caudatus	17	16-17	6	N/A	Grønfjorden, Svalbard	Lebedeva et al, 2018
Bivalvia						
H. arctica	49	150	25	N/A	Lancaster Sound	Atwell et al, 1998
Polychaeta						
Spionidae sp.	28	23-87	3	3-17	Bay of Fundy	Sizmur et al, 2013
Maldanidae sp.	60	101	16	70	Bay of Fundy	Sizmur et al, 2013
Chordata						
M. Scorpius	69-480	27-211	N/A	N/A	Canadian Arctic	van der Velden et al, 2013
		340±120		250±30	West Greenland	Rigét et al, 2007

4.3.1 Macroalgae

Concentrations of TotHg in lagoon macroalgae were low, but this was expected due to their roles as primary producers. Few studies from Arctic regions report TotHg concentrations in macroalgae, so comparison with our findings is difficult. Findings from a study in a polynya ecosystem in the Canadian Arctic (Clayden et al, 2015) showed that TotHg concentrations in *S.latissima* were up to 9 times higher than the values reported for Richardlaguna. However, we found that the TotHg concentration reported for this study was only slightly lower than *S.latissima* samples from Grønfjorden, Svalbard (Lebedeva et al, 2018). The variability in macroalgal Hg concentrations could be due to geographical differences, seasonal accumulation, site specific mechanisms, faster growth rates (due to enhanced productivity in lagoons) or a combination of all these factors (Coelho et al, 2005). Some macroalgal species tend to have bacterial colonies on their surfaces, which could act as an additional source of Hg and account for some of the variability found between studies. Macroalgae are important accumulators of Hg and can act an important vector for the incorporation of Hg to the base of estuarine food webs such as lagoons (Coelho et al, 2005). A study from Kongsjorden showed that some macroalgae species such as *Fucus sp.* can produce methylated forms of Hg in Arctic regions (Pongratz & Heumann, 1998). This species was found at Richardlaguna, so it could possibly act as a local source for producing bioavailable Hg. For the present study, some of the macroalgae species which were sampled had freely drifted into the lagoon. These specimens may play important roles in the transfer of Hg to the lagoon from the outer marine environment, especially during storms when there is the potential to transfer large masses of macroalgae to coastal environments. However, as we found uniformly low TotHg concentrations in macroalgae (both in FBM and DBM), this is likely not an important source of Hg to the lagoon environment.

4.3.2 Amphipoda

A wide range of TotHg concentrations has been reported for Arctic coastal amphipods, likely reflecting taxonomic differences, local and regional differences in contamination and ecological differences such as diet, growth rate and longevity (Legeżyńska et al, 2012; Legeżyńska et al, 2014). TotHg and MeHg in lagoon amphipods was lower than values reported for the Canadian Arctic (Clayden et al, 2015), but higher than the values found for amphipods sampled in Adventfjorden, Svalbard (Skogsberg, *unpublished data*), the coastal region of the Alkhornet bird cliffs, Svalbard (Finne, *unpublished data*) and at additional coastal sites in Isfjorden, Svalbard (Poste, *unpublished data*). TotHg in amphipods for the

present study were also generally higher than concentrations reported for coastal food webs in the eastern Canadian Arctic (van der Velden et al, 2013), although the %MeHg for the present study was lower.

4.3.3 Abundant benthic species

The TotHg and MeHg concentrations of benthic fauna presented for this study were variable across and within taxa. Interestingly we found that filter/suspension feeders (e.g. *H.arctica*) and some surface deposit feeders (e.g. *Terebellidae sp.* and *Maldanidae sp.*) had the highest TotHg concentrations, while predatory fauna (e.g. *Spionidae sp.* and *P.caudatus*) had the lowest concentrations. This indicates that benthic fauna that actively feed on suspended particles are experiencing a higher rate of TotHg accumulation compared to benthic fauna that are active predators. The highest MeHg concentrations was reported in the bivalve *H.arctica*, though no clear pattern was shown for MeHg concentrations for other benthic species in terms of feeding strategy. The higher range in $\delta^{13}C$ values for the lagoon bivalves may reflect that these organisms are feeding on a range of carbon sources (e.g. both tDOM and mDOM) and therefore have differential Hg in comparison to other lagoon benthic species. Given the less selective feeding strategy employed by bivalves (Jørgensen, 1996)), increased abundance of Hg-rich terrestrial particles could lead to increased Hg exposure and accumulation by these organisms. Though the tissue turnover times of stable isotopes in tissues and amongst species are different (Jardine et al, 2006), so it is difficult to comment on how representative the values of $\delta^{13}C$ and $\delta^{15}N$ are for 'snapshot' studies, which further complicates linking these data to Hg concentrations.

The range in Hg concentrations (TotHg and MeHg) for surface deposit feeders indicates that there are species-specific differences in Hg accumulation within this feeding guild. We found that of all the sampled polychaetes which are surface deposit feeders, *Maldanidae sp.* had the highest TotHg concentrations. This is consistent with observations from the temperate Bay of Fundy, where *Maldanidae sp.* had the second highest Hg concentrations of all sampled polychaetes behind *Capitellidae sp.* (a species which was not sampled in Richardlaguna) (Sizmur et al, 2013). *Maldanidae sp.* can feed deeper in the sediment profile (down to 20cm) (Jumars et al, 2015). This could in turn expose this species to heightened Hg concentrations, if there is a sufficient quantity of Hg stored in deeper sediments. Hg concentrations generally decrease with increasing sediment depth in Arctic ocean basins, although maximum Hg concentrations have been reported at sediment depths of 7-8cm (Gobeil et al, 1999). However, this may not be the case for Arctic coastal environments (including Richardlaguna)

as they have a different environmental setting compared to ocean basins, in that coastal environments experience a high degree of turbulent resuspension (due to influxes of freshwater from land) and bioturbation of sediments. This means that we would not expect a strong vertical change in sedimentary Hg based on temporal changes in inputs to the lagoon environment.

4.3.4 Sculpin

The TotHg concentrations found for sculpin in this study showed variability, with some individuals having concentrations higher than those reported in other Arctic areas (e.g. Rigét et al, 2007; van der Valden et al, 2013). The positive relationships we observed between TotHg and fish length and weight are consistent with a large number of studies related to contaminant accumulation in fish (Grieb et al, 1990; Dang & Wang, 2012; Julshamn et al, 2013; Bosch et al, 2016). There were no significant differences in TotHg muscle accumulation for sex or life stage, suggesting that the rate of Hg accumulation was the same for all sculpin.

Changes in diet may account for some of the TotHg variability as sculpin had differing $\delta^{15}N$ values and had a TL range of 3.7-4.2.However, this is a rather narrow range and is based on stable isotope and gut content analysis, which revealed that all sculpin sampled appeared to have quite similar dietary habits. Paired with the strong positive relationship between Hg and both length and weight, this suggests that age was the primary driver of variability in TotHg in sculpin, with long-term accumulation of Hg leading to higher TotHg in older fish (Gantner et al, 2010).

TotHg concentration in sculpin from Richardlaguna were below the toxicity threshold of 0.5 µg/g ww, were changes in biochemical processes, damage to cells and tissues, plus reduced reproduction can occur in fish (Dillon et al, 2010; Sandheinrich & Wiener, 2011). This was based on a conversion factor from dw to ww using 80% water loss in shorthorn sculpin muscle tissue (Harley et al, 2015).

4.3.5 Biomagnification of Hg through the lagoon food web

Sculpin from Richardlaguna had a higher mean TotHg BMF compared to American sand lance (*Ammodytes americanus*) feeding on gammarid amphipods, which were sampled in the Gulf of St. Lawrence, Canadian Arctic (Lavoie et al, 2010).

In a review paper by Lavoie et al (2013), TMS and TMF values for TotHg and MeHg biomagnification were evaluated globally for freshwater and marine food webs. Findings from that review paper showed that the TotHg TMS and TMF values reported for the present study were in line with the ranges reported for various ecosystems (including polar freshwater ecosystems, polar marine ecosystems and global coastal ecosystems) (Lavoie et al, 2013). Generally, the TotHg TMS for Richardlaguna was higher than the mean TMS values reported for temperate and tropical ecosystems (both freshwater and marine), which is consistent with the significant positive correlation between TotHg TMS values and latitude reported by Lavoie et al, (2013). This relationship is also the same for MeHg TMS values, however we do not report a MeHg TMS value for the present study as MeHg concentrations were only available for the most abundant benthic fauna.

Variability has been reported for TotHg TMS and TMF values across Arctic marine and freshwater food webs. The TotHg TMS value reported for Richardlaguna was higher than the TotHg TMS values reported in lacustrine and coastal food webs from the Canada Arctic (van der Velden et al, 2013; Clayden et al, 2015), but was generally lower compared to other Arctic marine food webs (e.g. Atwell et al, 1998, Campbell et al, 2005; McMeans et al, 2010). However, the TotHg TMF for Richardlaguna was generally higher or within the range of TMF's reported for other Arctic marine food webs (Jæger et al, 2009; van der Velden et al, 2013; Clayden et al, 2015). The variability in TMS and TMF values between Arctic marine and lacustrine food webs is likely due to several factors including physiochemical setting (e.g. pH, temperature, nutrients) which can influence growth rate, bioavailability of Hg, productivity which can influence trophic dilution of Hg, differential rates in excretion of Hg between organisms and complexity of food webs (Lavoie et al, 2013). The lower TotHg TMS value for Richardlaguna is probably linked to large variations in environmental parameters (e.g. salinity and temperature) across the year, which as has been reported for other Arctic lagoon ecosystems (e.g. Harris et al, 2017) and the enhanced productivity of lagoon systems (Knoppers, 1994; Dunton et al, 2006; Duck & da Silva, 2012; Dunton et al, 2012), leading to trophic dilution of Hg (Chen & Folt, 2005).

There are also several factors which must be taken into consideration when interpreting TMF values. As detailed in a review paper by Borgå et al, (2012), organism properties, characterisation of food webs with stable isotopes, ecosystem characteristics, spatial variation of contaminants across and within ecosystems, seasonal variation and chemical properties can all impact the calculation of TMF's and therefore the interpretation of a contaminant's

biomagnification potential. For example, calculation of TMF's is based on the regression between contaminants concentrations and TL, which assumes that the main route of contaminant exposure is dietary. For organisms such as fish and invertebrates this is not necessarily the case, as these organisms are also subject to direct uptake of contaminants from the surrounding water and sediments via exchange across respiratory surfaces (Borgå et al, 2012). The physiology of organisms is also important to consider when calculating TMF's. Alterations in ^{15}N can be impacted by growth rate, were in highly productive systems such as lagoons, the protein demand for the formation of new tissues is high and can lead to decreased enrichment factors of $\delta^{15}N$, while starvation can lead to increased enrichment (Hesslein et al, 1993). There may have been different $\delta^{15}N$ values in lagoon fauna if sampling had occurred at a different time of year (e.g. during the spring phytoplankton bloom), which could have led to a different TMF value for the Richardlaguna food web. It is therefore important to take these considerations into account when interpreting the biomagnification of contaminants through food webs when applying TMF's.

4.4. Climate change and coastal Hg cycling

Climate change is currently influencing the transport, speciation, distribution and cycling of Hg in the wider Arctic environment (Stern et al, 2012). Due to warming, reserves of previously deposited Hg are being released from storage media such as sea ice, snow, glaciers and especially permafrost, which stores a significantly large pool of Hg (Gordon et al, 2016; Schuster et al, 2018). This previously trapped Hg is subsequently being transported by meltwater and OM (Leitch et al, 2007), which could lead to increased Hg accumulation in coastal biota including in environments such as lagoons (Zhang et al, 2015).

Although we found that fauna in Richardlaguna tended to utilise marine carbon sources, future increases in terrestrial inputs will likely make terrestrial carbon more prevalent in coastal environments. This could increase Hg exposure to coastal fauna if transported particles are more enriched in Hg (as we found in this study) and there is a dietary shift which favours reliance on terrestrial carbon. Dietary changes which favour terrestrial carbon could impact Hg exposure in lagoon fauna as tDOM is often of lower quality and is first processed through the microbial loop, which adds further TL's in the food web and results in enhanced biomagnification of Hg (Karlsson et al, 2012; Karlsson et al, 2015; Jonsson et al, 2017; Creed et al, 2018). Lagoon fauna could also be directly exposed through consumption of Hg rich-terrestrial particles in fauna with less selective feeding strategies such as filter feeding (e.g. bivalves). This was observed in the present study, where we found that the bivalve *H.arctica*

fed on both terrestrial and marine carbon and also had one of the highest TotHg concentrations, as well as the highest MeHg concentration and %MeHg of all sampled benthic fauna.

Arctic coastal lagoons can experience long periods of ice cover throughout the year. With climate change leading to reductions in sea ice cover, these periods of time with prominent ice cover could be reduced and lead to an extension in the open water season. This could affect the cycling of Hg in these environments through enhanced exchange of Hg at the air/sea interface. There would be an increase in the quantity of Hg deposited from the atmosphere to surface waters, but also a higher rate of re-volatilisation of Hg to the atmosphere from surface waters. With less ice cover, there would also be greater rates of demethylation of organic Hg due to increased photochemical degradation from UV light (Stern et al, 2012). However, with the increased transport of terrestrial derived material, photo-demethylation of Hg could be reduced as higher loads of terrestrial particles lead to increased light attenuation, as has been shown in freshwater and coastal ecosystems (Poste et al, 2015; Klapstein et al, 2018). The combination of all these factors highlights the complexity of Hg cycling, so the impact of future climate change on Hg dynamics in lagoon systems is uncertain and therefore warrants further investigation.

4.5. Further work

Due to time constraints and currently ongoing method development, data on MeHg concentrations in lagoon samples is limited to only amphipods and abundant benthic organisms. This restricts our knowledge of how much bioavailable Hg is present in the lagoon environment and the degree to which MeHg is bioaccumulating and biomagnifying in the lagoon food web. In order to gain this insight, we are in the process of determining MeHg concentrations and %MeHg in remaining lagoon samples (water, sediment, POM, macroalgae, sculpin).

Due to the ‘snapshot’ nature of our sampling, our data on physiochemical conditions in the inlet streams, lagoon and marine environment are likely not representative of the year as a whole. Future work should therefore include a seasonal sampling strategy to account for environmental variations throughout the year, especially at times of the year with larger influxes of freshwater, terrestrially derived material and associated contaminants (e.g. spring/summer melting).

This study also lacks information on lagoon organisms which inhabit higher TL's, such as seabirds, Arctic char and marine mammals. Future sampling should include these organisms so that Hg (TotHg and MeHg) concentrations can be established, a more representative TMF can be calculated and further information on Hg concentrations in organisms can be established to investigate potential toxic effects and potential risk to humans can be evaluated (although the human health aspect is less important for Svalbard compared to other Arctic regions e.g. Canadian Arctic). Although, the use of these fauna does carry additional challenges, since their contaminant burdens can often reflect where they have migrated from as opposed to the environment of interest (in this case the lagoon).

It would also be beneficial to focus future studies on how lagoons with different catchment cover/degree of terrestrial influence, as well as differences in the degree of connectivity to the open ocean (e.g. open, semi-enclosed, closed) would influence the Hg cycling in these diverse ecosystems and how this would impact exposure and toxicity in lagoon fauna.

5. Conclusion

Despite being abundant coastal ecosystems, Arctic lagoon ecosystems are seldom studied. Climate change is leading to enhanced melting of media such as permafrost, which stores large quantities of Hg and is increased transport of tDOM and associated contaminants (like Hg) to Arctic coastal ecosystems. This increased loading from land to sea has potential implications on the food web structure and contaminant dynamics in Arctic coastal habitats such as lagoons. In this case study of Richardlaguna, a coastal lagoon on Svalbard, we found that stream inlets were the largest source of aqueous and particulate Hg to the lagoon. This highlights the significance of terrestrial inputs as transport vectors for Hg to Arctic coastal ecosystems. Despite being highly influenced by terrestrial inputs, lagoon fauna in Richardlaguna relied on marine derived energy sources, which contrasts with previous studies showing a high degree of reliance on terrestrial food sources in Alaskan coastal lagoons. Concentrations of TotHg and MeHg in lagoon fauna from the present study were generally higher than concentrations reported for other areas of Svalbard, although they were usually lower than concentrations found for other Arctic regions such as the Canadian Arctic. In order to characterise how ongoing environmental change may influence Hg dynamics in Arctic coastal ecosystems, future monitoring studies should focus on lagoon ecosystems as we have shown that these environments are potential hotspots for inputs of tDOM and Hg. It is therefore critical to understand processes controlling transport, fate and food web accumulation of Hg in Arctic coastal ecosystems in order to predict future responses to climate change and the potential for organisms to accumulate concentrations of Hg which could induce toxic effects.

6. References

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Appendices

Appendix A

Additional information on water sampling.

Parameter	Bottle Type	Preservation Agent	Sample Volume	Storage Conditions
TOC/TN	Amber Glass	H_2SO_4	100	Cool (4°C) and dark
TP	White Plastic	H_2SO_4	100	Cool (4°C) and dark
TotHg	White Plastic	HCl	250	Frozen (-20°C) and dark
MeHg	White Plastic	HCl	250	Frozen (-20°C) and dark
Nutrients	White Plastic	H_2SO_4	100	Cool (4°C) and dark
DOC	Amber Glass	H_2SO_4	100	Cool (4°C) and dark
DOM Char	Centrifuge Tube	N/A	50	Cool (4°C) and dark

Note: 1mL of preservation agent was pre-loaded into sample bottles when necessary, sample volume is expressed in mL, N/A = non-applicable.

Appendix B

Additional information on the number of individuals pooled and biometric measurements of amphipods and benthic infauna from Richardlaguna.

Date	Station	Collection Method	Taxon	Number Pooled	Weight (g)	Length (mm)
02/09/18	GN	Hand	*Gammarus sp.*	384	12.55	15-30
02/09/18	CB	Grab	*Brada villosa*	119	38.4	5-30
02/09/18	CB	Grab	*Mya sp.*	40	35.2	5-40
02/09/18	CB	Grab	*Chiridota laevis*	16	42.8	5-75
02/09/18	CB	Grab	*Polynoidea sp.*	50	10.4	4-35
02/09/18	CB	Grab	*Terebellidae sp.*	44	1.7	5-6
02/09/18	CB	Grab	*Spionidae sp.*	265	5.5	5-10
02/09/18	CB	Grab	*Macoma calcarea*	46	26.2	5-25
02/09/18	CB	Grab	*Maldanidae sp.*	35	3.7	10
02/09/18	CB	Grab	*Thyasira sp.*	67	1.4	3
02/09/18	CB	Grab	*Liocyma sp.*	88	6.4	6-7
02/09/18	CB	Grab	*Scoloplos armiger*	2	0.7	10
02/09/18	CB	Grab	*Priapulus caudatus*	32	2.4	5-10
02/09/18	CB	Grab	*Cylichna sp.*	3	0.4	5
02/09/18	CB	Grab	*Hiatella arctica*	15	22	10-30
02/09/18	CB	Grab	*Unknown gastropod*	1	0.4	12

Appendix C

Further information on filtration procedures for surface water samples from Richardlaguna. Filters marked with '*' were pre-weighed, while '^' were not pre-combusted.

Parameter	Filter	Pore size (µm)	Volume filtered (mL)	Storage Conditions
Chl *a*	GF/F^	0.7	300	Frozen (-20°C)
SPM	GF/F*	0.7	500-1000	Frozen (-20°C)
POM (SIA)	GF/F (PC)	0.7	500-1000	Frozen (-20°C)
PTotHg	QMA (PC)	2.2	500	Frozen (-20°C)
PMeHg	QMA (PC)	2.2	1000	Frozen (-20°C)

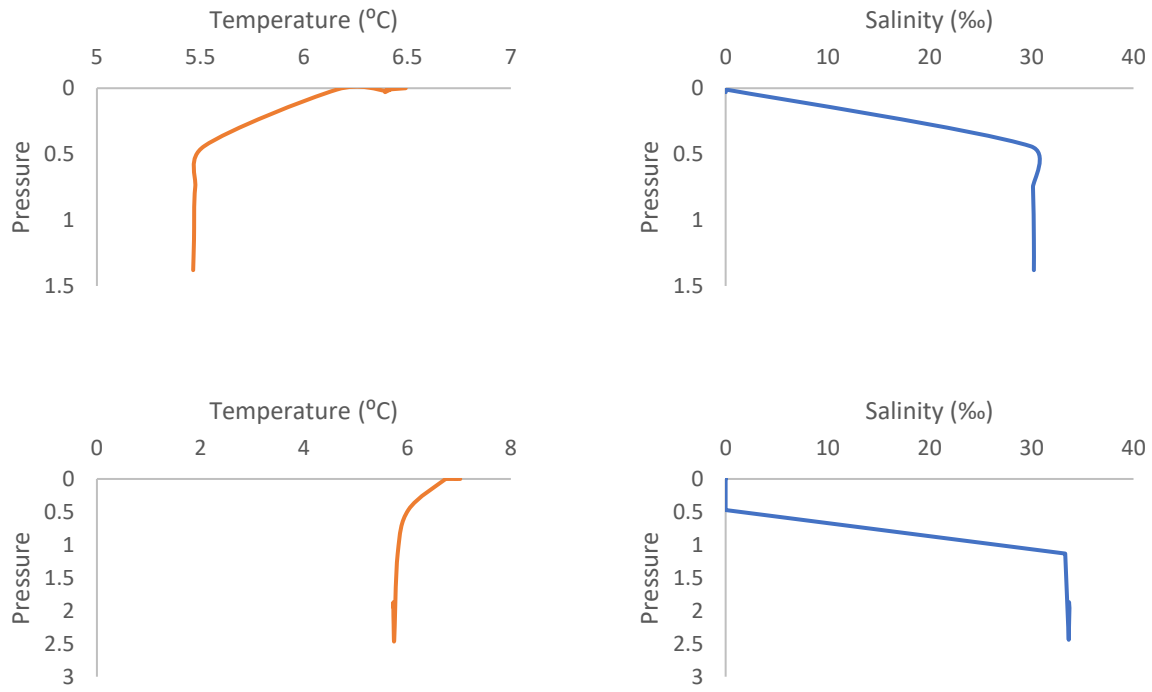
Appendix D

Additional information on sculpin sampled from Richardlaguna including biometrics and data from stomach content analysis.

Fish I.D.	Date	Sculpin Species	Length (cm)	Weight (g)	Sex	Stage	% Fill, Contents	Parasites
F224	02/09/18	Shorthorn	18.9	144.6	M	A	60, AM	Yes
F225	02/09/18	Shorthorn	13.7	66.5	M	A	40, AM	Yes
F226	02/09/18	Shorthorn	12.4	53.8	F	A	10, WD	Yes
F227	02/09/18	Shorthorn	15.3	91.7	M	A	50, WD	Yes
F228	02/09/18	Shorthorn	11.6	50.7	N/A	J	0	Yes
F229	02/09/18	Shorthorn	12.4	56.3	M	A	80, AM	No
F230	02/09/18	Shorthorn	14.6	88.9	F	A	0	No
F231	02/09/18	Shorthorn	11.3	36.7	N/A	J	10, WD	No
F232	02/09/18	Shorthorn	11.0	32.5	F	J	0	No
F233	02/09/18	Shorthorn	15.1	112.1	M	A	100, AM	No
F234	02/09/18	Shorthorn	15.8	108.0	N/A	A	0	No
F235	02/09/18	Shorthorn	11.4	45.2	F	J	0	Yes
F236	02/09/18	Shorthorn	16.5	110.2	N/A	A	40, WD	Yes
F237	02/09/18	Shorthorn	12.6	49.3	F	J	30, AM	Yes
F238	02/09/18	Shorthorn	15.3	74.6	N/A	A	0	Yes
F239	02/09/18	Shorthorn	11.8	42.5	N/A	J	10, WD	No
F240	02/09/18	Shorthorn	7.1	10.2	N/A	J	N/A	No
F241	02/09/18	Arctic Staghorn	9.4	23.5	F	A	N/A	No

Appendix E

Vertical profiles generated from CTD data for temperature (orange) and salinity (blue) plotted against pressure (depth) for the lagoon basin (upper) and lagoon outlet (lower).



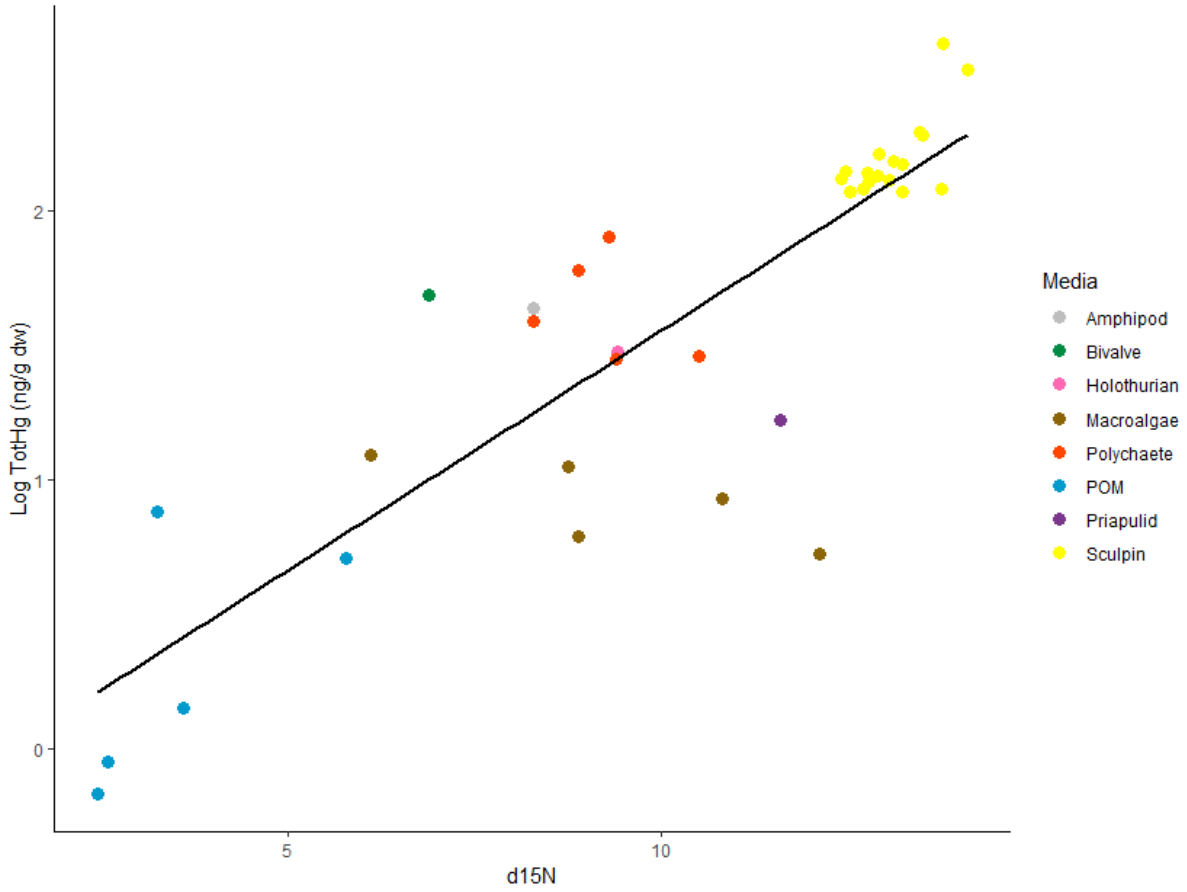
Appendix F

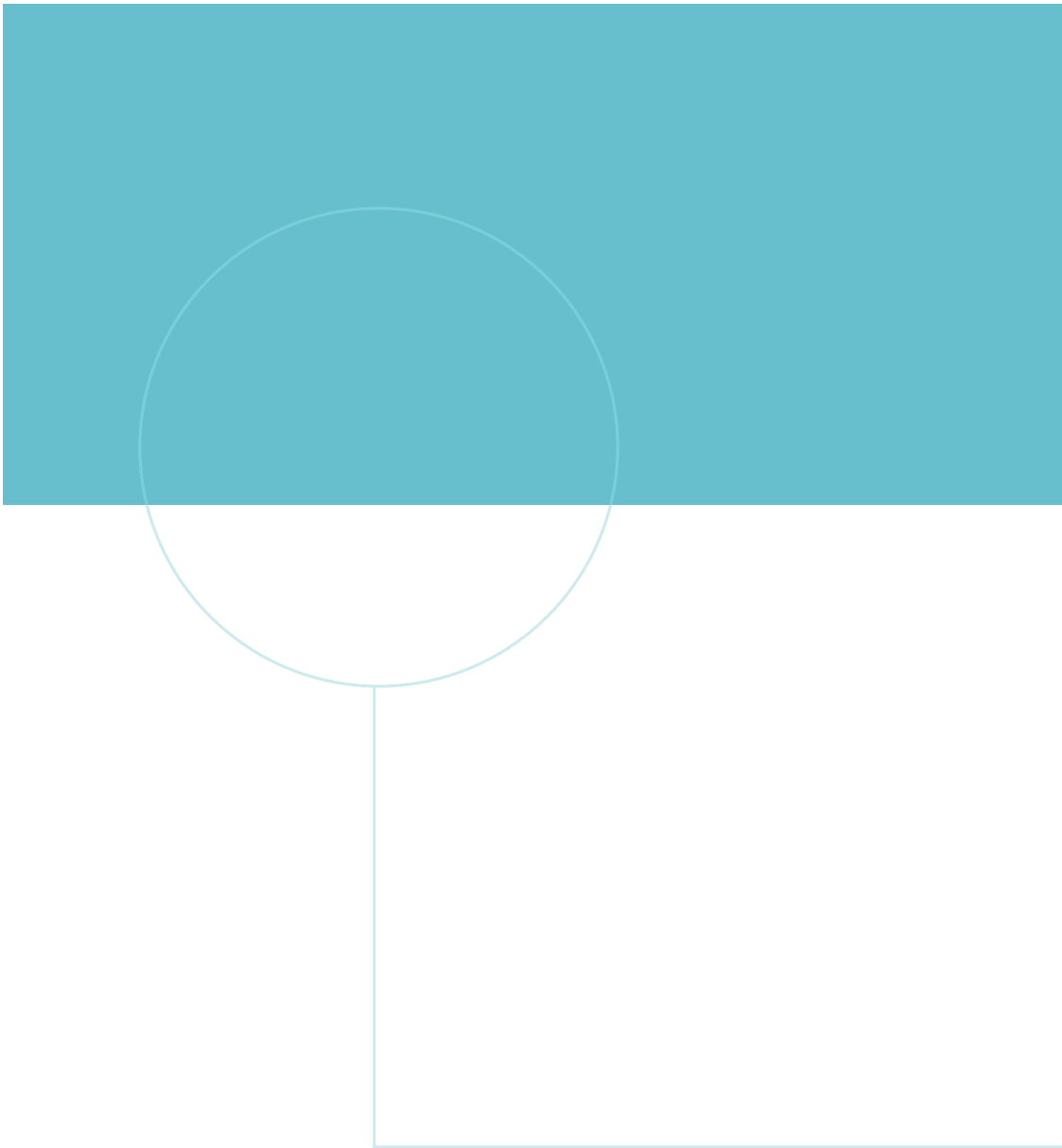
Results generated from the post-hoc multiple comparison Dunn's test. The level of significance is set to $p<0.05$, were significant relationships are denoted in bold and with '*'.

Comparison	Z-score	*p*-value (Unadjusted)	*p*-value (Adjusted)
Amphipod – Bivalve	-0.06	0.95	1.00
Amphipod – Holothurian	0.12	0.91	1.00
Bivalve - Holothurian	0.18	0.86	1.00
Amphipod - Macroalgae	0.64	0.52	1.00
Bivalve – Macroalgae	0.72	0.47	1.00
Holothurian – Macroalgae	0.49	0.63	1.00
Amphipod - Polychaete	0.03	0.98	1.00
Bivalve - Polychaete	0.11	0.92	1.00
Holothurian – Polychaete	-0.12	0.90	1.00
Macroalgae – Polychaete	-1.06	0.29	1.00
Amphipod – POM	0.96	0.34	1.00
Bivalve - POM	1.04	0.30	1.00
Holothurian – POM	0.81	0.42	1.00
Macroalgae - POM	0.55	0.58	1.00
Polychaete - POM	1.61	0.11	1.00
Amphipod - Priapulid	0.30	0.77	1.00
Bivalve - Priapulid	0.35	0.72	1.00
Holothurian – Priapulid	0.18	0.86	1.00
Macroalgae - Priapulid	-0.26	0.80	1.00
Polychaete – Priapulid	0.35	0.73	1.00
POM - Priapulid	-0.58	0.56	1.00
Amphipod - Sculpin	-1.18	0.24	1.00
Bivalve – Sculpin	-1.10	0.27	1.00
Holothurian – Sculpin	-1.34	0.18	1.00
Macroalgae – Sculpin	-3.85	0.12	**0.003***
Polychaete – Sculpin	-2.50	0.12	0.34
POM – Sculpin	-4.56	5.150278e-06	**0.0001***
Priapulid - Sculpin	-1.59	0.11	1.00

Appendix G

Linear relationship between Log TotHg (ng/g dw) and calculated trophic level (as derived from $\delta^{15}N$), which was used to calculate the trophic magnification factor of Richardlaguna.





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