Bioaccumulation of PCBs in Southern Resident Killer Whales in the Salish Sea

by

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Abstract

Chemical contaminants are a threat to Southern Resident Killer Whales (SRKW). The contribution of polychlorinated biphenyls (PCBs) in local sediments to the bioaccumulation of PCBs in SRKW was investigated. The temporal and spatial trends of concentrations of PCBs in sediment, Chinook salmon and SRKW were assessed. The half – lives of PCBs were estimated using a food web bioaccumulation model and the concentrations of PCBs in Chinook salmon and SRKW were estimated using Biota Sediment Accumulation Factors. There were no significant temporal declines in the concentrations of PCBs in sediment, Chinook salmon or SRKW as would be expected given the half – lives. The concentrations of PCBs in sediment could bioaccumulate to the levels observed in SRKW. Some similarities in the PCB congener composition were observed in sediment, salmon and SRKW. The results suggest that local environmental sources of PCBs in the Salish Sea could contribute to the PCBs observed in SRKW.

Keywords: Bioaccumulation; Southern Resident Killer Whales; polychlorinated biphenyls; Salish Sea

For my family and friends,

who supported me on my journey of career exploration and further education, and to Ryan Wickham, who always offered unconditional love and support throughout this process.

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List of Acronyms

BSAF	Biota Sediment Accumulation Factor
CCME	Canadian Council on Ministers of the Environment
CSAS	Canadian Science Advisory Secretariat
DFO	Fisheries and Oceans Canada
ECCC	Environment and Climate Change Canada
EIM	Environmental Information Management
NRKW	Northern Resident Killer Whales
PCB	Polychlorinated biphenyl
POP	Persistent organic pollutant
SARA	Species at Risk Act
SQG	Sediment Quality Guidelines
SRKW	Southern Resident Killer Whales
TL	Trophic level

Glossary

Bioaccumulation	The process where the chemical concentration in an aquatic organism exceeds the concentration in water due to chemical uptake through a variety of exposure routes. It is a combination of bioconcentration and biomagnification (Gobas and Morrison 2000).
Bioconcentration	The process where the chemical concentration in an aquatic organism exceeds the concentration in water due to absorption of the chemical from water (Gobas and Morrison 2000).
Biomagnification	The process where the chemical concentration in an organism exceeds the concentration in an organism's diet through dietary absorption by organisms at each trophic level of the food web (Gobas and Morrison 2000).
Biota – Sediment Accumulation Factor	The biota – sediment accumulation factor describes bioaccumulation in aquatic and sediment dwelling organisms relative to the chemical concentrations in sediment (Gobas and Morrison 2000).
Food web	The network of organisms and feeding relationships that control the trophic transfer of energy and contaminants.
Half – life	The half – life is the time required for the concentration of a chemical to reduce by 50% from the original concentration (US EPA 2015a).
Octanol water partition coefficient	The ratio of the chemical concentration of 1 – octanol and water. The octanol – water partition coefficient (K_{OW}) is used to represent how a chemical substance distributes between water and lipids in organisms (Gobas and Morrison 2000). Log K_{OW} is used to express hydrophobicity (Borgå et al. 2004).
Octanol air partition coefficient	The ratio of the chemical concentration of 1 – octanol and water in an octanol – water system at equilibrium (K_{OA}) (Gobas et al. 2009). The octanol – air partition coefficient is used to represent how a chemical partitions between lipids and air. It can be used to evaluate the potential of a chemical to bioaccumulate in food webs with air breathing organisms (Gobas et al. 2009).
Persistent organic pollutant	A class of organic chemicals substances that once released into the environment are persistent, bioaccumulative and toxic (Stockholm Convention 2004).
Sediment Quality Guidelines	A concentration of a chemical contaminant in sediment designed to protect all forms of aquatic life and all aspects of their aquatic life cycles during an indefinite period of exposure to the substances within sediments (CCME 2001).

Executive Summary

Chemical contaminants, including polychlorinated biphenyls (PCBs), have been identified as a threat to the health of the endangered Southern Resident Killer Whales (SRKW). PCBs have been linked to adverse health effects in marine mammals. PCBs continue to be the most prominent contaminant in sediment in coastal BC and a priority for monitoring due to the biological risk, despite being legacy contaminants. The Critical Habitat identified for the SRKW population is located within the Salish Sea in BC and Washington. The overall objective of this research is to investigate the contribution of local environmental sources of PCBs in the Salish Sea to the food web bioaccumulation of PCBs in SRKW. Several lines of inquiry will be explored to meet this objective. The research questions addressed in this project are: 1) What are the concentrations of PCBs in sediments of SRKW habitats, Chinook salmon and SRKW? How do they differ spatially and temporally? 2) Can the concentrations of PCBs in the habitat of SRKWs contribute significantly to the concentration of PCBs observed in Chinook salmon and SRKW? And 3) Does the composition of PCB congeners in sediment, Chinook salmon and SRKW provide clues on sources of PCBs to SRKW?

The contribution of PCBs stored in local sediment in the Salish Sea to the bioaccumulation of PCBs in Chinook salmon and SRKW was investigated in this research project. The results suggest that PCBs in the marine food web of SRKW originate to a significant extent from local environmental sources in the Salish Sea. Several lines of evidence provide support for this assertion including: i) the similarity of the temporal trends of the concentrations of PCBs in sediments, Chinook salmon and SRKW; ii) the presence of PCBs throughout the SRKW Critical Habitat at levels greater than the sediment concentrations recommended to be protective of SRKW; iii) the Biota Sediment Accumulation Factor (BSAF) modelling indicates the concentrations of PCBs in sediment could bioaccumulate to the levels of PCBs observed in SRKW; and iv) the similarity in the PCB congener patterns in sediments, Chinook salmon and SRKW. Given, the results suggest that there is an ongoing input of PCBs into the marine environment in the Salish Sea. If local environmental sources can be identified, then it may be possible to take management actions that will reduce the exposure of SRKW to PCBs.

Chapter 1.

Introduction

1.1. Threats to Southern Resident Killer Whales

The population of Southern Resident Killer Whale (*Orcinus orca*), SRKW hereafter, is facing imminent threats to their survival and recovery (DFO 2018c). The population size has been declining in the last two decades (Lacy et al. 2017) and there are currently only 74 individuals in the population. The SRKW population was listed as Endangered in 2003 under Canada's *Species at Risk Act* (S.C. 2002, c.29) (SARA) and was listed under the *US Endangered Species Act* in 2006. The three foremost anthropogenic threats to SRKW include prey availability, environmental contamination and physical and acoustic disturbance (DFO 2017a). The Government of Canada has outlined a recovery strategy and detailed action plans to mitigate these threats as required by SARA (DFO 2008, 2011; Ford et al. 2017). There were 99 recovery measures identified in the Recovery Strategy including 21 related to environmental contaminants.

High levels of environmental contaminants, including polychlorinated biphenyls (PCBs), have been identified as a threat to the health of SRKW. A recent global assessment of PCB effects on the long term viability of killer whale populations suggests that PCBs alone have the potential to lead to population collapse (Desforges et al. 2018). PCBs are legacy contaminants that were banned from use in North America and Europe in the 1970's (Addison and Ross 2000). PCBs continue to be the most prominent contaminant in sediment in coastal BC and a priority for monitoring due to the biological risk (Alava et al. 2012a; Morales-Caselles et al. 2017). Recent monitoring has shown that the concentration of PCBs in SRKW samples has not significantly declined since the mid – 1990's (Guy 2018) despite these contaminants having significantly declined in harbour seals from the Salish Sea (Ross et al. 2013b). This observation suggests that there continue to be inputs into the marine environment that supports the SRKW food web bioaccumulation of these particular group of persistent organic pollutants (POPs). It is critical to identify whether local environmental sources of PCBs contribute to the PCB levels observed in SRKW and their main prey, Chinook salmon, to reduce health risks from exposure to PCBs. As migratory species, Chinook salmon can accumulate PCBs from local coastal marine habitats and the offshore marine environment. If local environmental sources of PCBs in the Salish Sea contribute significantly to the PCBs transferred via the food web to SRKW, then it may be possible to reduce PCB inputs to the SRKW food web and habitat through local initiatives.

1.2. Objective and Research Questions

The overall objective of this research is to investigate the contribution of local environmental sources of PCBs to the food web bioaccumulation of PCBs in SRKW. Several lines of inquiry will be explored to meet this objective. The research questions addressed in this project are:

1. What are the concentrations of PCBs in sediments of SRKW habitats, Chinook salmon and SRKW? How do they differ spatially and temporally?
2. Can the concentrations of PCBs in the habitat of SRKWs contribute significantly to the concentration of PCBs observed in Chinook salmon and SRKW?
3. Does the composition of PCB congeners in sediment, Chinook salmon and SRKW provide clues on sources of PCBs to SRKW?

Chapter 2.

Background

2.1. Legislation and policies to protect Southern Resident Killer Whales in Canada

The Government of Canada has committed to working with all levels of government, Indigenous peoples, industry and environmental stakeholders to implement the *Species at Risk Act* (SARA) (DFO 2018d). One of the purposes of SARA is "*to provide for the recovery of wildlife species that are extirpated, endangered or threatened as a result of human activity*" (DFO 2011). Recovery is defined as "*the process by which the decline of an endangered, threatened or extirpated species is arrested or reversed, and threats are removed or reduced to improve the likelihood of the species' persistence in the wild*". A Recovery Strategy was developed in 2008 by the Government of Canada that sets goals, objectives and main activities required to conserve Resident Killer Whales (DFO 2008). The final recovery strategy was published in 2011. The three main threats, including physical and noise disturbance, reduced availability and quality of prey and environmental contaminants, are addressed in the four principal objectives of the recovery strategy (DFO 2017a). The objective in the recovery strategy that is applicable to environmental contaminants is: "*ensure that chemical and biological pollutants do not prevent the recovery of Resident Killer Whale populations*" (DFO 2011).

The federal ministers of Fisheries and Oceans (DFO) and Parks Canada Agency are responsible under SARA for preparing Action Plans to implement the Recovery Strategy (DFO 2017a). The 2017 Action Plan for the Northern and Southern Resident Killer Whales outlines how the objectives identified in the recovery strategy will be achieved. Broad Strategy 4 of the Action Plan outlines six different approaches to ensure chemical and biological pollutants do not prevent recovery of Resident Killer Whales (DFO 2017a). Each approach describes recovery measures, the priority, timeline and partners who will assist with addressing some aspect of the main three threats. There are 21 specific measures related to environmental contaminants.

In the 2018 Budget, the Government of Canada introduced $167 million for a 5 - year Whale's Initiative to address threats to Canada's endangered whales (SRKW, North

Atlantic Right Whales, *Eubalaena glacialis*, and St. Lawrence-Estuary Beluga, *Delphinapterus leucas*). This initiative includes commitments to increase monitoring and improve understanding of sources and impacts of contaminants on whales and their prey and introduce stronger contaminant control measures by 2020 (Transport Canada 2018). In May 2018, the Ministers of Fisheries and Oceans and Environment and Climate Change Canada determined that the SRKW population is facing imminent threats to the survival and recovery of the population (DFO 2018a). The Ministers recognized that intervention was required for the continued survival and eventual recovery of the SRKW population (DFO 2018a). The list of threats to the SRKW population was expanded beyond the three key threats previously identified to include oil spills, incidental mortality in fisheries and ship strikes (DFO 2018c). In May 2018, the government took action to increase prey availability by reducing the total Chinook salmon caught by fisheries by 25 – 35% and closing commercial and recreational fisheries in key whale foraging areas (Transport Canada 2018).

2.2. Southern Resident Killer Whales

2.2.1. SRKW Distribution and Critical Habitat

The coastal waters of British Columbia (BC) and Washington are inhabited by three ecotypes of killer whales (*O. orca*), Transients, Offshore and Resident Killer Whales. The ecotypes have different diets, genetics, behaviour and morphology (Ford et al. 1998). Resident Killer Whales consist of Southern and Northern distinct populations that have overlapping habitat range and feeding habitats, but are culturally, genetically and acoustically distinct (DFO 2008, 2018c). There are three pods (J, K and L) within the SRKW Population.

The habitat range of SRKW extends from Monterey Bay in California to Chatham Strait in southeastern Alaska (Ford et al. 2017). Most sightings of SRKW occur in late spring to early fall within Haro Strait, eastern Juan de Fuca Strait and the Strait of Georgia (Ford et al. 2017). This 3,390 km^2 area was identified as Critical Habitat in Canada in the 2008 recovery strategy because SRKW spend a significant portion the year in this area, especially when foraging for Chinook salmon (DFO 2017b; Ford et al. 2017) (Figure 2.1). Recently, an additional 5,025 km^2 area off southwestern Vancouver Island has been proposed as Critical Habitat for SRKW due to observations of all three

SRKW pods using this area (Figure 2.1). Under the SARA (2002) section 2(1)Critical Habitat is defined as "…*habitat that is necessary for the survival or recovery of a listed wildlife species and that is identified as the species' Critical Habitat in a recovery strategy or action plan for the species*" (DFO 2011). The US government has also identified a 6,630 km ^{2} area in Puget Sound and the surrounding area in the Juan de Fuca Strait as Critical Habitat under the *US Endangered Species Act* in 2006 (Ford et al. 2017) (Figure 2.1). The Critical Habitat for SRKW in the US and Canada fall within the Salish Sea. The Salish Sea is a transboundary area encompassing the Strait of Georgia, Juan de Fuca Strait and Puget Sound that is rich in marine life supporting approximately 3,000 species (PSF 2016). This entire area is especially important to all three SRKW pods for foraging during Pacific salmon migration (DFO 2018c).

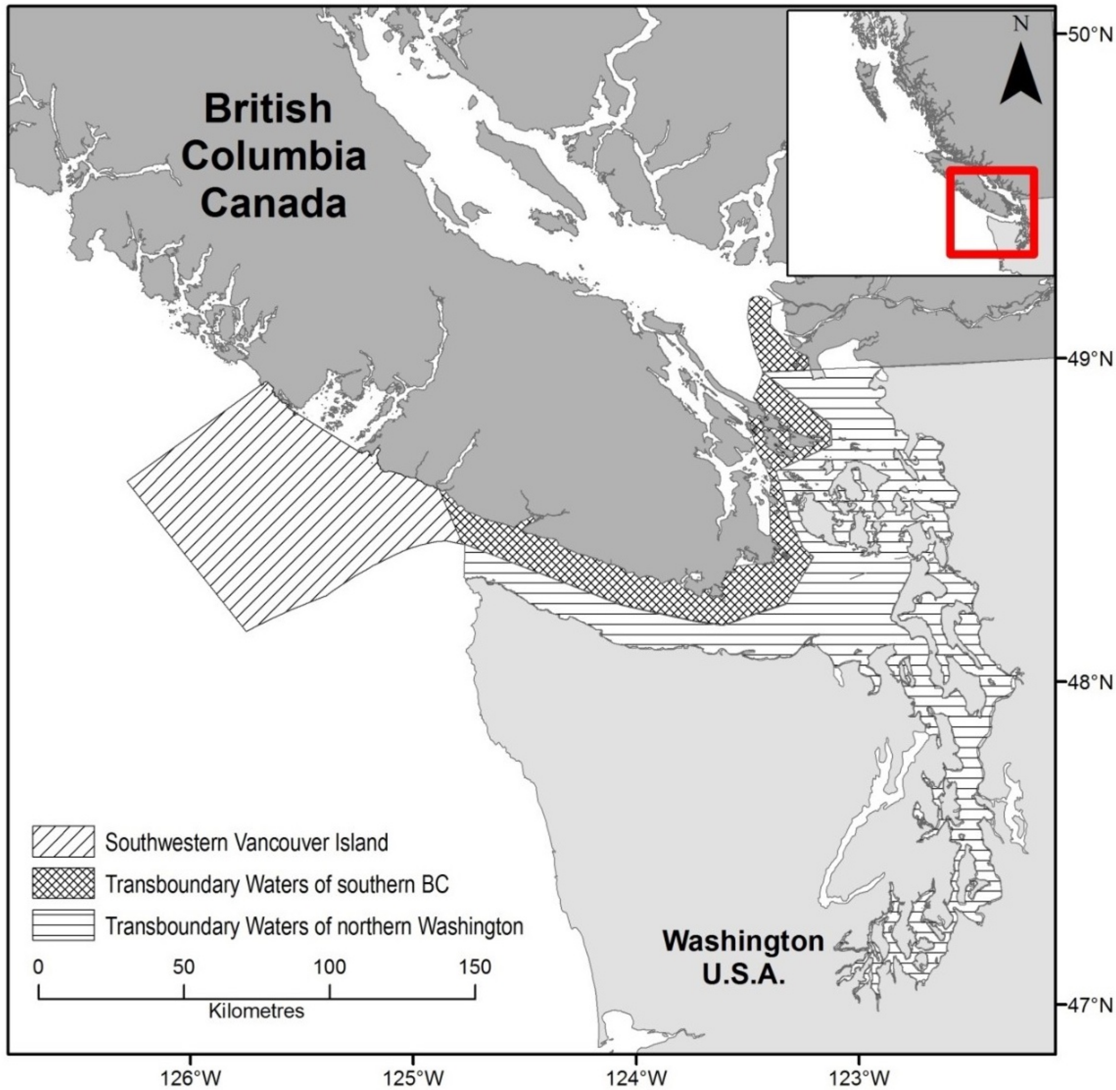

Figure 2.1 The Critical Habitat for Southern Resident Killer Whales in the transboundary waters (i.e. Salish Sea) of southern British Columbia (BC, Canada) and northern Washington (USA). The proposed future Critical Habitat is the area off of southwestern Vancouver Island.

Source: Southern Resident Killer Whale Imminent Threat Assessment (DFO 2018c).

2.2.2. SRKW Food web

The Resident Killer Whale food web varies spatially and temporally (Lachmuth et al. 2010; Alava et al. 2012a). Organisms at the same trophic level generally have similar levels of PCB contamination. The trophic guilds most relevant to bioaccumulation and transfer of PCBs include phytoplankton and algae, zooplankton and filter feeding invertebrates and benthic detritivores (i.e., amphipods, crabs, shrimp and polychaetes), forage and predatory fish (Lachmuth et al. 2010) (Figure 2.2). Pacific Salmon (*Oncorhynchus* spp.), particularly Chinook salmon (*Oncorhynchus tshawytscha*), are the preferred prey of Resident Killer Whales during the summer and fall (Ford et al. 1998; Ford and Ellis 2006). Coho salmon (*O. kisutch*) are the second highest consumed salmonid species (15% of diet) (Ford et al. 2016). Chinook salmon are the preferred Pacific salmon species because of they are larger and have higher lipid content (Ford and Ellis 2005). Also, SRKW can consume Chinook salmon year – round because Chinook salmon spend more of their lifecycle in coastal waters than other salmon species. SRKW forage on Chinook salmon in the southern Strait of Georgia, Puget Sound and Juan de Fuca Strait from May to September (Hanson et al. 2010; Ford et al. 2017). Puget Sound becomes an increasingly important foraging area in October and November when SRKW prey on Chinook salmon and migrating Chum salmon (DFO 2018c). Therefore, this study has focussed on Chinook salmon that originate from natal streams or rivers in these areas and spend time in these areas. SRKW primarily feed on Chinook salmon from the Fraser River system (80% of diet), especially the South Thompson stock (Ford et al. 2017).

Chinook salmon are anadromous fish that inhabit both freshwater and marine habitat during their life history and they are migratory species. Chinook salmon migrate throughout the Pacific Ocean (Figure 2.3). However, detailed information on migratory routes of individual populations is challenging to find. In addition to varying migration routes, Chinook salmon have evolved into two distinct types, “stream – type” and “ocean – type”, that spend a different amount of time in coastal waters compared to offshore waters (Hope 2012). The stream – type of Chinook salmon spend more time in freshwater before they migrate as yearlings to coastal waters followed by 2 to 4 years of offshore migration to the Gulf of Alaska and Northern Pacific Ocean (Healey 1991; Hope 2012). The stream – type are caught in high seas fisheries. Whereas the ocean – type of Chinook salmon spend more time as juveniles in estuaries than in freshwater before migrating along the coast (Hope 2012). The ocean – type are more common in sheltered coastal waters than the stream – type of Chinook salmon (Healey 1991a). Chinook salmon stocks from northern BC are mixed stream and ocean – type, whereas Chinook salmon from southern BC and Puget Sound are predominantly ocean – type (Healey 1991a). Generally, Chinook salmon return to the Fraser River to spawn at around 4 to 5 years old (Healey 1991) and Chinook salmon return to Puget Sound streams after 2 to 4 years in the ocean (Essington et al. 2011).

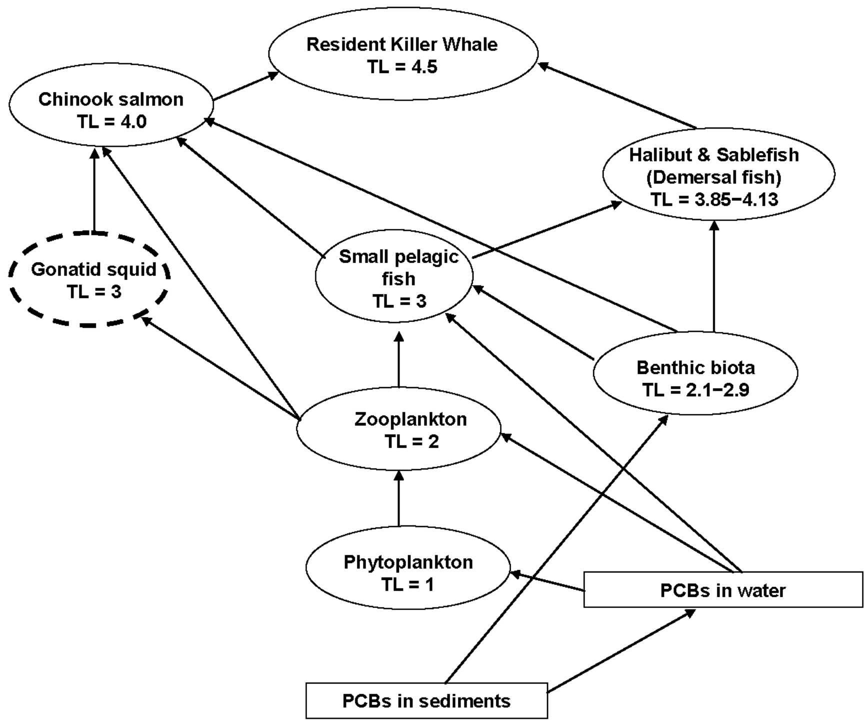

Figure 2.2 Diagram of PCB entry into the marine food web of Resident Killer Whales and trophic levels (TL) of the species in the food web (adapted from Alava et al. 2012a).

Reprinted with permission from Alava, J. J., Ross, P. S., Lachmuth, C., Ford, J. K. B., Hickie, B. E., & Gobas, F. A. P. C. (2012). Habitat-Based PCB Environmental Quality Criteria for the Protection of Endangered Killer Whales (Orcinus orca). *Environmental Science & Technology*, *46*(22), 12655–12663. Copyright (2012) American Chemical Society.

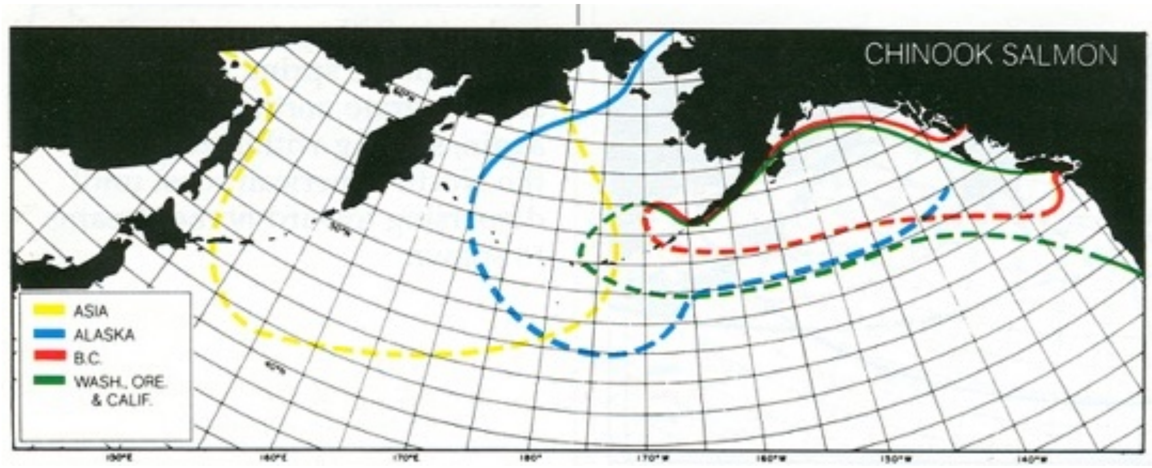

Figure 2.3 Chinook salmon migration in the Pacific Ocean. The migration route of populations originating from BC is indicated in red.
Source: Salmon facts – Pacific salmon (DFO 2018b).

2.3. Polychlorinated Biphenyls

2.3.1. PCB Production and Regulation

PCBs are a group of synthetic organic chemical compounds comprised of 209 different forms or congeners (ATSDR 2000). PCB congeners consist of one to ten chlorine or hydrogen atoms attached to two benzene rings (biphenyl) (ATSDR 2000). They were produced in commercial mixtures of PCB congeners, such as Aroclors (Megson et al. 2015). PCBs were introduced in the 1930's as lubricants and liquid insulators used primarily in electrical equipment and later used in a variety of household and industrial products (ECCC 2017b).

The phasing out of PCBs in North America and Europe began in the 1970's. In the US, the manufacture, processing and distribution of PCBs was banned in 1976 under the *Toxics Substances Control Act* (Washington State Department of Ecology 2015). In Canada, the manufacture, import and sale of PCBs was banned in 1977 (ECCC 2017a). PCBs are defined as persistent, bioaccumulative and toxic within the Stockholm Treaty on Persistent Organic Pollutants (POPs) (UNEP 2001). In Annex A, of the Stockholm Treaty on POPs, PCBs are identified as a contaminant scheduled for elimination (Porta 2002). Parties are required to stop using existing equipment that contains or is contaminated with PCBs by 2025 (UNEP 2016a). In Canada, the PCB Regulations (SOR/ 2008-273) outline how this deadline will be met by ending use of equipment and products containing PCBs, including light ballasts, transformers (Government of Canada 2008).The PCB Regulations are intended to "*protect the health of Canadians and the environment by preventing the release of polychlorinated biphenyls (PCBs) to the environment, and by accelerating the phasing out of these substances*" (Government of Canada 2008). As of 2015, it was estimated that approximately 14 million tons of PCBs still needed to be eliminated worldwide (UNEP 2016b).

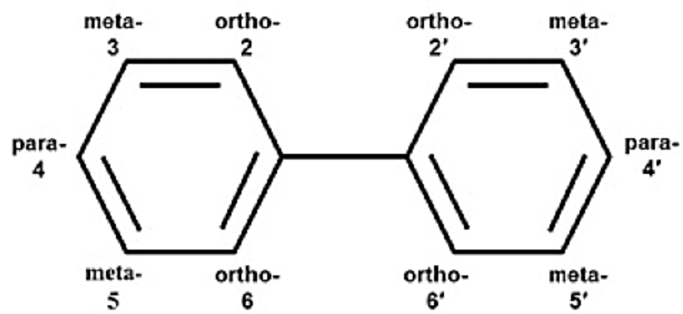

Figure 2.4 Chemical structure of PCBs. The labels of meta, ortho, and para indicate the location of the chlorine atoms. The numbers indicate possible locations of chlorine atoms on the benzene rings (ATSDR 2000).

Source: ATSDR. 2000. Toxicological Profile for Polychlorinated Biphenyls (PCBs). Chemical and Physical Information. https://www.atsdr.cdc.gov/toxprofiles/tp.asp?id=142&tid=26.

2.3.2. Sources of PCB Contaminants

The sources of PCBs in the Salish Sea include localized industrial activities, sediments, long range atmospheric transport from global sources and biological transport by migratory species, including salmon (Ikonomou et al. 2002; Johannessen et al. 2008b; Christensen et al. 2005; West et al. 2008; Noël et al. 2009; Alava et al. 2012a). After PCBs were banned in Europe and North America, the use and improper disposal of PCBs in developing countries led to ongoing global atmospheric distribution of PCBs (Addison and Ross 2000). In North America, PCBs can be released into the environment from leaks or releases from electrical transformers containing PCBs, disposal of consumer products containing PCBs in landfills, burning wastes in incinerators, run off from contaminated soils and effluents (CCME 2001; US EPA 2015b). PCB contamination in Puget Sound has been attributed to historic industrial activity (Ross et al. 2004) and proximity to highly developed cities (West et al. 2017). The Strait of Georgia is bordered by highly populated areas in Greater Vancouver, and has received discharge from industrial activities including pulp mills, mining and municipal wastewater effluents (Johannessen et al. 2008b). It is estimated that wastewater effluent contributes less than 10% of PCBs stored in sediment in the Strait of Georgia (Johannessen et al. 2015).

2.3.3. PCBs in the Marine Environment and Biota

PCBs are ubiquitous in the marine environment and are found in biota all over the globe (Kelly et al. 2007; Blasius and Goodmanlowe 2008; Noël et al. 2009; Alava and Gobas 2012; Frouin et al. 2013; Desforges et al. 2018). Once PCBs enter the environment, they primarily partition into the organic carbon fraction of sediment (Lachmuth et al. 2010; Johannessen et al. 2015). PCBs are primarily removed from sediment through the process of burial. PCB half - lives in sediment depend on many factors, including the chemical properties of the specific PCB congener, with estimates of half - lives ranging from several years to over 100 years (Sinkkonen and Paasivirta 2000).

There is constant exchange between environmental compartments (e.g. water, air, sediment, biota) through environmental and biogeochemical processes such as volatilization, sediment burial and organic carbon cycling, as shown in Figure 2.5 (Johannessen et al. 2008b; Johannessen et al. 2015). Fish can become contaminated with PCBs through gill uptake of PCBs in water (bioconcentration) and dietary uptake from their food (biomagnification). Sediment dwelling invertebrates can take up PCBs directly from the sediment. Plankton can take in PCBs from the water (Frouin et al. 2013). The concentrations of these contaminants can increase at each trophic level of the food chain (Kelly et al. 2007; Gobas and Arnot 2010; Alava et al. 2012a). PCBs accumulate in lipids in biota due to their chemical properties, particularly the high octanol - water partition coefficient (K_{OW}) (Lachmuth et al. 2010). PCBs have been shown to be toxic to fish at low concentration and to have adverse health effects in humans and wildlife (ECCC 2017b).

The Canadian Council for Ministers of the Environment (CCME) have set an interim sediment quality guideline (ISQG) for marine sediments of 21.5 μg/kg dry weight that is designed to protect benthic invertebrates (CCME 2001). The SQG were not specifically developed to account for adverse effects in higher trophic levels that are subject to bioaccumulation and biomagnification in their food web (CCME 2001; Alava et al. 2012a). This has been identified as a particular concern for aquatic organisms at higher trophic levels (Alava et al. 2012a; Arblaster et al. 2015). The SQG has been found to be too high to protect most wildlife species and humans consuming seafood products.

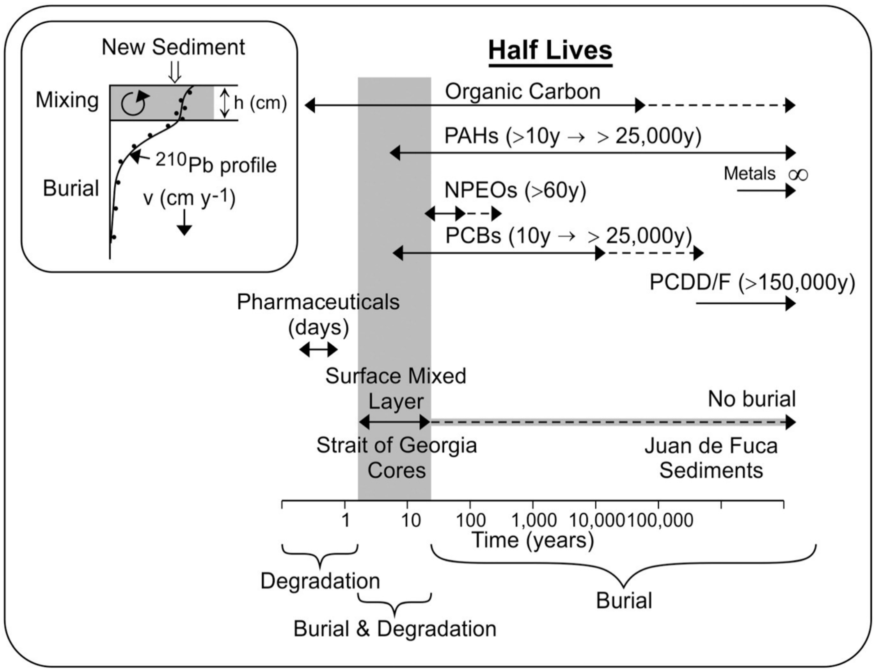

Figure 2.5 A schematic diagram comparing the importance of sediment burial and degradation for removing contaminants from the Strait of Georgia (Johannessen et al. 2015).

Reprinted from Science of the Total Environment, 508, Sophia C. Johannessen, Robie W. Macdonald, Brenda Burd, Albert van Roodselaar, Stan Bertold, Local environmental conditions determine the footprint of municipal effluent in coastal waters: A case study in the Strait of Georgia, British Columbia, 228-239., Copyright (2015), with permission from Elsevier.

2.4. PCBs in Killer Whales

2.4.1. Accumulation of PCBs in Killer Whales

There are a number of factors that influence PCB accumulation in killer whales, including age, sex and dietary preference (specifically trophic level) and life history exposure (Ross et al. 2000). As long lived species, killer whales can accumulate PCBs throughout their lifespan and pass the contaminants onto their offspring (Ross 2006b; Desforges et al. 2018). Monitoring studies have detected PCBs in killer whales since the 1990’s (Addison and Ross 2000b; Ross et al. 2000; Ross 2006b; Krahn et al. 2007). As of 2012, the concentrations of PCBs were available for approximately one third of the SRKW population (Mongillo et al. 2012). There have been 136 PCB congeners detected in killer whales (Addison and Ross 2000; Ross et al. 2000). Higher chlorinated congeners generally account for the highest portion of ∑PCB while lower chlorinated congeners are absent or present at low levels (Ross et al. 2000). The position and number of chlorine atoms will affect the persistence, solubility, bioaccumulative potential, toxicity and volatility of the PCB congener (Mongillo et al. 2016). Through metabolism, dioxin – like[1] PCBs are eliminated by killer whales, but the mono-ortho PCBs and other globular (nonplanar) - structured POPs are not eliminated at the same rate (Tanabe et al. 1988; Ross 2006a). This has been attributed to low cytochrome P450 activity in cetaceans (Tanabe et al. 1988).

The marine mammal-eating transient killer whales have been found to have higher concentrations of PCBs than fish - eating Resident Killer Whales because the transient killer whales consume prey that are at higher trophic levels (Addison and Ross 2000; Ylitalo et al. 2001). Within Resident Killer Whales, SRKW have been found to have higher PCB contamination than the Northern Resident Killer Whales (NRKW) (Addison and Ross 2000; Ross et al. 2000). Although both SRKW and NRKW feed preferentially on Chinook salmon, SRKW consume Chinook salmon closer to more contaminated habitats and when the salmon have reduced lipids as they are returning to spawn (Cullon et al. 2009). It is estimated that SRKW consume 4 to 6.6 times more PCBs than NRKW on a body weight basis (Cullon et al. 2009). With a life expectancy of females of up to 90 years and a life expectancy of approximately 50 years for males (Hickie et al. 2007), some older individuals in the Resident Killer Whale population have been alive since before PCBs were banned in North America. However, there is no method to directly assess how the PCB exposure history relates to the concentrations of PCBs measured in Resident Killer Whales (Hickie et al. 2007).

2.4.2. Modelling of PCBs in Resident Killer Whales

Different modelling approaches have been used to improve understanding of PCB exposure and resulting PCB contamination over the lifespan of Resident Killer

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[1] Dioxin – like PCBs include MO PCBs 105, 114, 118, 123, 156, 157, 156 and 189 and NO PCBs 77, 81, 126, and 189 (Ross et al. 2000)

Whales (Hickie et al. 2007; Mongillo et al. 2012). Individual based modelling has been conducted to estimate historical, current and future Concentrations of PCBs for individual whales within the SRKW population based on their life history characteristics and prey contaminant concentrations (Mongillo et al. 2012). Of the three SRKW pods, J pod was predicted to have the highest concentrations of PCBs (Mongillo et al. 2012). The J pod resides in the Salish Sea throughout the year (Ford et al. 2017).

A food web bioaccumulation model has been used to estimate the Concentrations of PCBs in Resident Killer Whales based on the PCB sediment concentrations in their Critical Habitat and other locations within their habitat range (Lachmuth et al. 2010; Alava et al. 2012a). This model described the relationship between concentrations of PCBs in sediments, Chinook salmon and resident killer whales (Alava et al. 2012a). The results of food web bioaccumulation modelling using sediment quality criteria and tissue residue guidelines for PCBs in Canada predicted total Concentrations of PCBs in Chinook salmon and Resident Killer Whales would exceed toxicity threshold concentrations known to cause adverse health effects in marine mammals (Alava et al. 2012a). The current environmental sediment quality criteria for PCBs have been found to be too high to be protective of Resident Killer Whales.

2.4.3. Health effects of PCBs in Southern Resident Killer Whales

In all marine mammals, it is challenging to determine causal relationships between contaminant exposure and adverse health effects. This is because there are natural confounding factors, multiple toxic chemicals, other anthropogenic influences on health (e.g. fishing, noise and climate change) and ethical, legal and logistical challenges associated with sampling large mammals (Ross 2006b). There is no direct evidence of the PCB related health effects on Resident Killer Whales (Hickie et al. 2007). To understand the health risk of PCB contamination to killer whales, scientists have referenced toxicological effects in other marine mammals and used various modelling approaches (Mos et al. 2010; Alava et al. 2012; Desforges et al. 2018). PCBs and other lipophilic contaminants have been shown to adversely affect reproduction, immune function and endocrine function of marine mammals in coastal regions close to industrial activity (Tabuchi et al. 2006; Mos et al. 2006; Buckman et al. 2011; Desforges et al. 2016; Peñin et al. 2018). A weight of evidence approach that used results collected in laboratory and field studies of marine mammals suggested that the concentrations of PCBs found in killer whales were sufficient to be a toxicological risk to killer whale populations in BC (Ross 2000). PCBs have been found to alter the abundance mRNA related to the health of killer whales, specifically increased gene expression in the aryl hydrocarbon receptor, thyroid hormone α receptor, estrogen α receptor, interleukin 10 and metallothionein 1 (Buckman et al. 2011). In a recent population viability analysis of SRKW, it was stated that the impact of PCBs on calf survival is the only health effect with sufficient data to use in predictions of the effects of PCBs on population level demographic rates in SRKW (Lacy et al. 2017). Ultimately, the health risk to Resident Killer Whales from exposure to PCBs depends on many factors including age, sex, dietary preferences and calving order (Hickie et al. 2007).

2.5. PCBs in Chinook Salmon

The accumulation of PCBs by Chinook salmon in is a key component of the food web bioaccumulation of PCBs by Southern Residents. PCB exposure and accumulation in Chinook salmon will vary depending on traits and movement patterns between freshwater and marine habitats. Pacific salmon accumulate POPs when they are in the ocean where they spend most of their life and where the majority of their growth occurs (Cullon et al. 2009; Ross et al. 2013b). Puget Sound Chinook salmon accumulate approximately 96% of PCBs during their time in marine habitats rather than in freshwater as juveniles (O'Neill and West 2009). Further, the ocean – type and stream – type of Chinook salmon will have different food sources and exposure to PCBs because they spend time in different marine habitats (e.g. coastal waters versus offshore). The ocean – type spend more time in coastal waters that are potentially closer to anthropogenic sources of contamination (Carlson and Hites 2005). Chinook salmon from BC, Puget Sound in Washington and Oregon, that are likely ocean – type, have been shown to have higher concentrations of PCBs compared to Chinook from Alaska, that are likely stream – type (Carlson and Hites 2005).

The concentrations of PCBs in Chinook salmon collected in British Columbia and Washington have been found to exceed the dietary PCB threshold of 8 μg/kg that is estimated to protect 95% of a killer whale population based on an adapted PCB adverse effects threshold for marine mammals (Hickie et al. 2007; Cullon et al. 2009; Arblaster et al. 2015). There has been limited monitoring of the concentrations of PCB in Chinook salmon from the Fraser River which are the main population of Chinook salmon consumed by SRKW. Therefore, the research conducted on Chinook salmon from Puget Sound can be used to improve our understanding of the accumulation of PCBs by Chinook salmon populations in the Salish Sea. The concentrations of PCBs in Puget Sound Chinook salmon have been observed to be higher than the concentrations of PCBs observed in other West Coast populations of Chinook salmon (Missildine et al. 2005; O'Neill and West 2009) and farmed salmon (Carlson and Hites 2005). It has been suggested that sources of PCB contamination may be present within Puget Sound or along the migratory route of Puget Sound Chinook salmon in the Pacific Ocean that Chinook salmon from coastal areas of Washington are not exposed to (Missildine et al. 2005). Given that a portion of Chinook salmon remain Puget Sound residents, the elevated concentration of PCBs in these salmon may be attributed to contamination in Puget Sound (O'Neill and West 2009). It is a challenging task to identify where Pacific salmon uptake contaminants because they are exposed to various sources of contaminants along their migratory route and these migratory routes are variable (Ross et al. 2013a).

Chapter 3.

Methodology

3.1. Question 1: What are the concentrations of PCBs in sediments of SRKW habitats, Chinook salmon and SRKW? How do they differ spatially and temporally?

3.1.1. Sample Collection

Samples of sediment, Chinook salmon and SRKW were collected and analyzed by other researchers as part of environmental monitoring programs and research studies in BC and Washington. The concentrations of PCBs in sediment, Chinook salmon and SRKW are publicly available or are currently part of ongoing research. This includes unpublished data that have been provided for use in this project.

Sediment

British Columbia Strait of Georgia and SRKW Critical Habitat

Sediment samples were collected as part of several monitoring and research programs in the southern Strait of Georgia and SRKW Critical Habitat in Canada over the past few decades. In the 1990's and early 2000's sediment samples were collected in the Strait of Georgia, Victoria Harbour and Vancouver Harbour, but the years that samples were collected could not be verified, so these observations have not been included in this study (Arblaster 2012; Arblaster et al. 2015). From 2002 to 2007, sediment samples were collected in BC as part of the Strait of Georgia ambient monitoring program which was a collaboration between DFO, Natural Resources Canada and Metro Vancouver (Wright 2008). These samples were collected using corer samplers (Pouliout Box and Pederson) and grab samplers (Shipek and Eckman). Additional details on the sample collection methods used in the Strait of Georgia ambient monitoring program are available in previous publications (Johannessen et al. 2008b; Wright 2008; Grant et al. 2011). From 2010 to 2017, sediment samples were collected by Environment and Climate Change Canada (ECCC) staff during environmental monitoring surveys at Disposal at Sea sites and nearby locations that have not been subject to loading from dredging materials (Ross et al. 2011a, 2012; ECCC 2017c). The sediment samples used in this study were collected in the Strait of Georgia from a Coast Guard research vessel using Shipek or Smith – McIntyre grab samplers (Ross et al. 2011b). In addition, sediment samples were collected throughout coastal BC in 2015 and 2016 during Phase 1 of the "PollutionTracker" program of the Ocean Wise Coastal Ocean Research Institute (CORI) program (Ocean Wise-CORI, 2018; http://pollutiontracker.org/contaminants/pcbs/). For this project, the concentration of ∑PCB in sediment was compiled for the sampling stations in the Strait of Georgia and surrounding waterway off the southeast coast of Vancouver Island and around the lower mainland. The "PollutionTracker" samples were collected from a small vessel using a Petit Ponar grab sampler (Ocean Wise-CORI 2018).

Washington SRKW Critical Habitat

Sediment samples were collected by the Washington State Department of Ecology as part of the Puget Sound Ecosystem Monitoring Program Long Term Sediment Component (PSEMP) and Puget Sound Assessment and Monitoring Program (PSAMP) (Washington State Department of Ecology 2018a). Marine sediment quality data are publicly available for download from the Environmental Information Management System (EIM) (Washington State Department of Ecology 2018a). To obtain the data for this study, the EIM database was searched using the selection criteria of parameter name of "PCB" and salt/ marine sediment data. Survey data were available from 1989 to 2016 at the time of data compilation for this project.

Sediment samples were collected throughout northern Washington near the San Juan Islands, Eastern Strait of Juan de Fuca, Admiralty Inlet, Whidbey Basin, Central Puget Sound, South Puget Sound and Hood Canal at designated stations (Washington State Department of Ecology 2018b). The purpose of the environmental monitoring was to identify long - term ecosystem change in Puget Sound (Washington State Department of Ecology 2018b). The sediment samples were collected using a modified van Veen sediment grab at designated monitoring locations (Dutch et al. 2009).

Chinook salmon

Chinook salmon samples were collected in 2000 and 2014 in BC coastal waters. The current study focussed on these Chinook salmon because SRKW have been found to primarily consume Chinook salmon from the Fraser River (Ford et al. 2017). The adult

Chinook salmon samples from 2000 were collected from Johnstone Strait and the mouth of the Fraser River (Cullon et al. 2009). The details of the sample collection methods are available in the original publication (Cullon et al. 2009). The 2014 Chinook salmon head samples were collected off of Southern Vancouver Island when the salmon were returning from sea (Guy 2018). The stock ID, timing and specific collection location are not available.

Southern Resident Killer Whales

The concentration of PCBs in SRKW blubber and tissues were compiled by SFU graduate student Jayda Guy from previously published studies and recent sampling in 2015 (Krahn et al. 2007; Ross et al. 2013). The new SRKW samples were collected and analyzed in 2015 in partnership with DFO and the US National Oceanic and Atmospheric Administration (Guy 2018). The concentrations of PCBs in SRKW samples were used in a risk assessment as part of Jayda Guy's Master's thesis (Guy 2018).

3.1.2. Chemical Analysis

Sediment

British Columbia SRKW Critical Habitat and Strait of Georgia

The concentrations of PCB congeners in sediment samples collected from 2002 to 2007 were analyzed using High Resolution Gas Chromatography – High Resolution Mass Spectrometry (HRGC - HRMS) according to United States Environmental Protection Agency (US EPA) EPA 1668 method (Grant et al., 2011; Johannessen et al., 2008). Values were reported on a dry weight basis. Further details are provided in previous publications (Johannessen et al. 2008b; Wright 2008; Grant et al. 2011). The 2010 samples were analyzed for PCBs by the DFO Laboratory for Expertise in Aquatic Chemical Analysis (LEACA), in Sidney, BC (Ross et al. 2011b). The concentration of PCBs in the sediment samples collected from 2011 to 2017 by ECCC were measured using HRGC/HRMS in accordance with the United States Environmental Protection Agency (US EPA) 1668 method by several labs including ALS Global, Analytical Ltd. and Maxxam Analytics .

Sediment samples were analyzed for congener - specific PCBs and reported as individual congeners, co – eluting congeners and homologues (Ross et al. 2011b; ECCC

2017c). The 2015 and 2016 sediment samples were analyzed for 209 PCB congeners. Further details on analysis will be presented in future publications (Ocean Wise-CORI, 2018).

Washington SRKW Critical Habitat

The concentration of PCBs in the sediment samples collected in Washington were analyzed by gas chromatography – electron capture detector (GC – ECD) in accordance with EPA 8082A (Dutch et al. 2018). The concentrations of PCBs in sediment were analyzed for 21 individual PCB congeners and 9 Aroclor mixtures (Dutch et al. 2018). The specific analysis methods reported in the EIM database include the Organochlorine Pesticides and Polychlorinated Biphenyls (PCBs) by GC, combined method, PCBs by Gas Chromatography with Electron Capture Detection (SW8082A, Revision 1) and Organochlorine Pesticides (SW8081B, Revision 2) (Washington State Department of Ecology 2018a). Analyses were conducted by laboratories accredited by the State of Washington including the Department of Ecology's Manchester Environmental Laboratory (MEL) (Dutch et al. 2018). The export from the EIM database stated the data were verified and assessed as usable for "Formal Study Report" and "Peer- Review Study Report" (Washington State Department of Ecology 2018a). Values were reported on a dry - weight basis.

Chinook salmon

The Chinook salmon samples collected in 2014 were analyzed for PCBs using HRGC - HRMS at Axys Analytical Ltd. in Sidney, British Columbia using HRGC - HRMS instrumentation in accordance with the US EPA 1668 protocol (Guy 2018). The Chinook samples collected in 2000 were analyzed using HRGC - HRMS at the DFO Regional Contaminants Laboratory (Cullon et al. 2009). Further details on the contaminant analysis methods are available in the original published study (Cullon et al. 2009). All analysis were conducted in accordance with the US EPA 1668 protocol (Guy 2018).

Southern Resident Killer Whales

The SRKW samples collected in 2015 were analyzed for PCBs using HRGC - HRMS at Axys Analytical Ltd. in Sidney, British Columbia (Guy 2018). The samples collected between 1993 and 2009 were analyzed by HRGC/HRMS with the exception of one 2004 analysis conducting using LC GC/MS (Guy 2018). All analysis were conducted in accordance with the US EPA 1668 protocol (Guy 2018).

3.1.3. Data Preparation for Analyses

Sediment

British Columbia Strait of Georgia and SRKW Critical Habitat

For this study, the results of multiple environmental monitoring surveys and research projects conducted were compiled with different standards of reporting PCB concentration values. When the concentration of PCBs in sediment was reported as sum of PCB congeners in the original publications, these values were used. In the Strait of Georgia, dataset collected between 2003 – 2007 congeners that were undetectable in greater than 30% were excluded (Grant et al. 2011). Further details on data preparation are available in the original publications (Johannessen et al. 2008a; Wright 2008).

The results of ECCC environmental monitoring surveys from 2010 to 2017 were compiled in an Excel database by ECCC staff from survey and lab reports and were provided for use in this study (Dr. Justin Lo, *personal communication,* 2017). The BC sediment dataset included PCB concentration values that were indicated as below the detection limits of the chemical analysis method or censored data. PCB congeners that were indicated as below the detection limit in greater than 70% of samples were excluded from subsequent analyses in this study. The rationale for this approach was to exclude PCB congener concentration data with high uncertainty. After excluding the low quality data, three different methods were compared to deal with non – detects in the PCB congeners that were detectible in at least 30% of samples. The methods included substituting non - detects with zero, substituting non - detects with half the detection limit and ignoring all values below the detection limit or not detected. This approach is similar to approach used in other studies to reduce the influence of congeners with high rates of non – detection on the ∑PCB concentration (Alava et al. 2009; Ross et al. 2011b; Desforges et al. 2014). The PCB sediment concentration datasets produced using the three substitution methods described above were log 10 transformed for subsequent analyses in this study except where specifically stated. Student t – tests were conducted to determine if there was a significant difference between the mean total concentrations of PCBs in sediment produced using each of the three substitution methods. The t - test results indicated that there was not a statistically significant difference in the ∑PCB concentration produced from any of these methods. The normality of the concentrations of ∑PCB in sediment in BC was assessed by plotting the sample quantiles of the log transformed data against the theoretical quantiles of a standard normal distribution (Normal quantile – quantile plot) and by plotting histograms (Appendix B). Based on the results of these assessments, it was determined that using substitution with half the detection limit provided the best fit to a normal distribution. The dataset produced by substituting non – detects with half the detection limit was used in all subsequent analyses of the PCB sediment concentration in BC. The concentrations of PCBs in sediment that were reported as ng/kg dry weight were converted to pg/g dry weight.

Washington SRKW Critical Habitat

Marine sediment quality data were compiled for each survey by the Washington State Department of Ecology staff according to standardized procedures and entered into the EIM database. According to the Quality Assurance Monitoring Plan for the Puget Sound Sediment Monitoring Program, non - detects in all sediment chemistry will be reported at the reporting limits for the sample. Any summary statistics would be estimated using regression order statistics or Kaplan-Meier censoring techniques (Dutch et al. 2018). Therefore, methods of dealing with non - detects were not compared in this study for the Washington sediment dataset downloaded from the EIM website. The concentrations of PCBs in sediment that were reported as μg/kg dry weight were converted to pg/g dry weight.

Chinook Salmon and Southern Resident Killer Whales

The concentration of PCBs in Chinook salmon and SRKW blubber samples were compiled by Jayda Guy and the data were published in her Master’s thesis (Guy 2018). The detection limit was substituted if the concentration of PCBs was below the detection limit. The results of this approach was compared to using half the detection limit and assuming the concentration of PCBs was zero (Guy 2018). The approach for dealing with non – detects was found to not have a significant effect on the results (Guy 2018).

The concentrations of PCBs in Chinook salmon were lipid normalized using the lab measured lipid content for each fish which ranged from 2.08% to 15.08% (Guy 2018). The concentration of ∑PCB for the 2015 SRKW samples were lipid normalized using the lab measured lipid content of 64.3%. The concentration of PCBs in SRKW samples collected earlier were lipid normalized using lipid content obtained from previously published literature which ranged from 9.6% to 64.3% (Guy 2018).

3.1.4. Spatial Trend Analysis of PCBs in Sediment

Mapping

Maps were created using ESRI ArcGIS software to show the location where individual sediment samples were collected in BC and Washington (ESRI 2016a). The NAD 83_HARN projection was used. To produce the map, all the raw survey data from 2010 to 2017 were imported into the GIS software and the latitude and longitude coordinates for each sample were displayed as points. This time period was chosen because there was a large sample size for each year and sample coordinates were reported for all locations where sediment samples were collected in BC which was not the case for earlier years. For consistency, the same sample time period was selected for the dataset from Washington SRKW Critical Habitat.

The SRKW Critical Habitat in Canada and US were also included on the map to show how the sampling locations overlapped with the Critical Habitat. The polygons were created from shape files provided by DFO (Robin Abernethy (DFO), *personal communication*, 2017).

Spatial Interpolation

The spatial analysis tools in ESRI ArcMap were used to interpolate the concentration of PCBs in sediment in the Salish Sea (Strait of Georgia, Juan de Fuca Strait and Puget Sound) and produce a map showing the spatial variation of concentration of PCBs throughout the Salish Sea. Specifically, the Inverse Distance Weighting (IDW) Raster Interpolation method from the ArcMap 3D spatial analysis toolbox was used to estimate a continuous surface of concentrations of PCBs in sediment and produce a map layer of these values (ESRI 2016b). IDW is a deterministic interpolation method where the value of a given variable is estimated based on the values of the variable at nearby areas. In this case, the concentrations of PCBs in areas that were not measured were estimated by using the observed concentrations of PCBs in sediment observed at the point sample locations. The IDW method of interpolation was chosen for this study because it assumes the measured values are spatially correlated (ESRI 2016b). In this study, spatial correlation was assumed to be an appropriate assumption to use to describe the relationship between concentrations of PCBs in sediment and distance. Also IDW is a widely used interpolation methods because it is fast to compute and relatively easy to interpret compared to geostatistical interpolation methods (Lu and Wong 2008).

The estimated or interpolated concentration of ∑PCB in sediment were generated in ArcMap based on the known observed concentrations of ∑PCB in sediment collected from 2010 to 2017 throughout the Salish Sea. The interpolated value ($Z\ (x_0)$) was calculated using 1 to *n* measured concentrations of PCBs where x_i is the *i*th measured value and h_{ij} is the distance between the measured data point and the location of the interpolated data point (Equation 1) (Bhunia et al. 2018). A variable search method was used to find input values for the interpolation. A minimum of 10 sample points were used to predict a concentration value up to a maximum of 0.5 map units away. If these conditions were not met, then no value was predicted for that location. The power value (β) controls how much influence the input values have based on the distance from the output point (ESRI 2016c). In this study, the default power value of 2 was selected. Therefore, the influence or weights assigned to each measured input concentration of PCBs are inversely proportional to the squared distance between the observed sample point location and the estimated point (ESRI 2016b). The influence of different power values was not assessed in this study.

$$Z\ (x_0) = \frac{\sum_{i=1}^{n} \frac{x_i}{h_{ij}^{\beta}}}{\sum_{i=1}^{n} \frac{1}{h_{ij}^{\beta}}} \qquad (1)$$

To produce a map of the spatial variability in concentrations of PCBs in sediment within the Salish Sea, the interpolation was set to constrain the analysis to the boundary of the Salish Sea. Specifically, a map layer of the Salish Sea was used to set the processing extent for input values and as a raster analysis mask to limit the output estimates of concentration of ∑PCB in sediment to only this geographic region. The resulting interpolated map layer of a smooth surface of concentration of ∑PCB in sediment in the entire Salish Sea was displayed using a colour scale. The interpolated results were displayed using a colour gradient by changing the symbology in the layer properties. The interpolated values of concentrations of ∑PCB in sediment were stretched along a color gradient with the minimum value (100 pg/g dw) of concentrations of PCBs shown in blue and the maximum value of concentrations of PCBs shown in red (60,000 pg/g dw). The colour gradient was demarcated at fixed interval widths of 10,000 pg/g of PCBs in sediment.

3.1.5. Temporal Trend Analysis of PCBs in sediment and biota

Sediment

The sediment monitoring data were divided into two groups, including BC and Washington, to assess overall temporal trends. The BC dataset included samples collected between 2002 and 2017 in the Strait of Georgia and the SRKW Critical Habitat Canada. The Washington dataset included samples collected between 1989 and 2016 in the US SRKW Critical Habitat in Puget Sound and Juan de Fuca Strait. However, to match the time period for the BC dataset, only the data collected from 2002 to 2016 was used in the temporal analysis. Concentrations of total PCBs (∑PCB) in sediment were calculated as the sum of all measured individual congeners and co-eluting congeners for all individual sediment samples where possible. If only ∑PCB were reported (e.g. samples collected in BC before 2010), then the reported concentrations of ∑PCB in sediment were used. All data were previously treated for non – detects where applicable as described previously in the Sample Preparation section.

To assess how the concentration of ∑PCB in sediment changed over time in BC (Strait of Georgia and Canada SRKW Critical Habitat) and Washington (US SRKW Critical Habitat), a linear regression approach was used. All analyses and plots were completed in R statistical software (R Core Team 2017). Simple linear regression of the log transformed concentrations of ∑PCB in sediment by sample collection date and by year were completed. Diagnostic plots were produced to assess whether the linear model met the assumptions of linear regression including normality of residuals and equal variance of residuals (Appendix B). The slope of the regression line and its associated *p* – value was used to determine whether there was a change in concentration per unit time and the significance of the trend. The coefficient of determination r - squared (R^2) value was used to assess the proportion of variance in concentrations of PCBs that is captured by the linear regression by year (Fox 1997;

Nakagawa et al. 2017). The R^2 is often used in biological studies to understand the sources of variation in biological data (Nakagawa et al. 2017). Although there was some evidence of temporal auto – correlation, a mixed linear regression model with years as a random effect was not used because the mixed linear model does not have a standard R^2 value associated with the slope model that can be used to evaluate the model fit (Nakagawa and Schielzeth 2013).

The geometric mean of the concentration of ∑PCB in sediment and standard deviation were calculated for BC and Washington for each year. The geometric mean reduces the bias and high variability introduced by using data from environmental monitoring studies that may focus on locations that are known to have elevated concentration of PCBs in sediment (Lachmuth et al. 2010). To further investigate the difference in the mean concentration of ∑PCB in sediment, Analysis of Variance (ANOVA) and Tukey Honest Significant Difference (Tukey HSD) tests were completed using the log 10 transformed concentrations of ∑PCB in sediment. The ANOVA test identified if there was any difference in any of the annual mean concentration of ∑PCB in sediment. When there was a difference in at least one year, a Tukey HSD test was used to simultaneously compare the concentration of ∑PCB between all years. The test results showed whether the mean concentration of ∑PCB in sediment was significantly different from the mean concentration of ∑PCB in sediment observed in other years.

The percent change by year in concentrations of ∑PCB in sediment by year was compared to the half - life time of PCBs in sediment in the Strait of Georgia and Puget Sound. In the Strait of Georgia the half - life of PCBs in the surface mixed layers is estimated to be 10 years (+/- 8 years) (Lachmuth et al. 2010). This estimate was calculated from sedimentation rates of 0.3 to 3 cm/year and surface mixed layers 4 to 25 cm deep (Lachmuth et al. 2010). At two sample sites in Central Puget Sound, the sedimentation rate was estimated through radiometric dating to be 1.3 cm/year (+/- 0.1) and 2.1 +/- 0.3 cm/year (Brandenberger et al. 2008). The sediment surface mixing depth was 10 – 20 cm (Brandenberger et al. 2008). Based on the values reported by Brandenberger et al. (2008), the half – life of PCBs in Puget Sound sediment was calculated to be approximately 6.5 years (+/- 3.1 years) or ranging from 3 to 10 years.

There are several caveats to consider when interpreting the results of the analysis of the temporal trends in concentration of PCBs in sediment. The temporal trends found in this study are subject to the sample locations where the original data were collected that particular year. However, aggregating the data for an entire geographic region increases the sample size and decreases the influence that the measured PCB concentration at any one location would have on the mean ∑PCB concentration. It is assumed in the temporal analysis that the sediment concentrations of PCBs are representative of the PCB concentration in the sediment throughout the area. However, samples were not taken at the same location each year and there are limited samples in earlier years, so the observed concentrations are representative of the sediment samples collected in any given year and provide an approximation of the concentrations throughout the area. Also replicate sediment samples were collected in close proximity to other samples during a single survey and there could be repeat sampling at each location in multiple years, but all were treated as individual samples in this study.

Chinook Salmon

The lipid normalized concentrations of ∑PCB in Chinook salmon samples were used for analysis of the temporal trends. A simple linear regression was fitted to the log transformed concentrations of ∑PCB in Chinook salmon collected in 2000 and 2014. As described above for sediment, diagnostic plots were produced to assess whether the assumptions of linear regression were met (Appendix B).

The geometric mean concentration of ∑PCB in Chinook salmon was calculated by taking the mean of the log 10 transformed concentrations of lipid normalized ∑PCB measured from samples collected in 2000 ($n = 12$) and 2014 ($n = 7$). The temporal trend analysis and the geometric mean concentration of ∑PCB in Chinook salmon must be interpreted with caution because of the limited data available.

Southern Resident Killer Whales

The SRKW dataset provided by Jayda Guy were subset into adult male, adult female and juvenile whales prior to conducting subsequent analyses in this study. The age of “adult” was defined as a minimum age of sexual maturity reported in published literature. The age of adult females was set at 12 years and the age of adult males was 10 years (Olesiuk et al. 1990; DFO 2008). There was a total of 40 samples for SRKW including adult females ($n = 15$), adult males ($n = 19$ with 4 repeat observations of the same individuals) and juveniles (n = 6). The lipid normalized concentrations of PCBs in tissue and blubber SRKW samples were used.

To assess how the concentration of ∑PCB in adult female and male SRKW changed over time from 1993 to 2015, simple linear regressions were completed. As described above for sediment, diagnostic plots were produced to assess whether the assumptions of linear regression were met (Appendix B). The slope of the regression lines was used to determine the change in concentration per year and the *p* – value was used to assess whether the change over time was statistically significant.

The geometric mean of concentrations of ∑PCB in adult female and adult male SRKW were calculated by taking the mean of the log 10 transformed concentrations of PCBs for each year. ANOVA and Tukey's HSD tests were conducted to compare the annual mean concentration of ∑PCB in SRKW samples and determine which years the mean total concentrations of PCBs in SRKW were significantly different.

3.2. Question 2: Can the concentrations of PCBs in the habitat of SRKWs contribute significantly to the concentration of PCBs observed in Chinook salmon and SRKW?

3.2.1. Comparison of concentrations of PCBs in sediment and biota

To illustrate the biomagnification relationship between the environmental media and biota, the concentration of ∑PCB in sediment, Chinook salmon and SRKW were compared. The geometric mean concentration of ∑PCB in in sediment in BC was calculated from all samples collected in the Strait of Georgia and the Canadian SRKW Critical Habitat from 2010 to 2017 by ECCC. Compiling data from multiple years increased the sample size and decreased the influence of the individual locations sampled in any given year. To enable a comparison between concentrations of PCBs in sediment and biota, the geometric mean ∑PCB in sediment was converted to lipid equivalent units. The geometric mean concentration of ∑PCB in sediment was normalized by organic carbon content of 1.50% in sediment (upper SD 2.89% and lower SD 0.78%). Subsequently, the organic carbon normalized geometric mean concentration of ∑PCB in sediment was divided by a factor of 0.35 to convert to lipid equivalent values

(Seth et al. 1999). The factor of 0.35 represents the correlation between the organic carbon/ water partition coefficient (K_{OC}) and the K_{OW} (Seth et al. 1999).

The geometric mean concentration of ∑PCB in sediment in US SRKW Critical Habitat was calculated form all samples collected in Juan de Fuca Strait and Puget Sound in Washington. The organic content for each sediment sample was not included in the dataset obtained from the Washington State Department of Ecology EIM database. Therefore, values for the organic carbon contents of the sediments in Puget Sound were obtained from a previous Washington State Department of Ecology report (Pelletier and Mohamedali 2009). The geometric mean of the organic carbon contents in the top 2 cm was calculated from the average organic carbon content reported for individual sampling sites. The value of 1.58% (upper SD is 2.10% and lower SD is 1.19%) was used to normalize the concentration of ∑PCB in sediment in the US SRKW Critical Habitat. However, it is acknowledged that since the organic carbon content was obtained from a different survey, the value may not be representative of the actual organic content of the sediment samples where the concentrations of PCB in sediment were measured. To convert the organic carbon normalized concentration of ∑PCB in sediment to lipid equivalent values, the geometric mean was divided by a factor of 0.35 (Seth et al. 1999).

Due to the low sample sizes for Chinook salmon and SRKW, all available data were used to calculate the geometric mean concentration of ∑PCB. The geometric mean was calculated from the lipid normalized concentrations of ∑PCB in adult male SRKW samples collected between 1993 and 2015 and from adult female SRKW samples collected between 1996 and 2015. The geometric mean concentration of lipid normalized ∑PCB in Chinook salmon was calculated from the samples collected in 2000 and 2014.

3.2.2. BSAF estimated concentrations of PCBs in Chinook salmon and SRKW

BSAF model calculations

The Biota Sediment Accumulation Factor (BSAF) characterizes the relationship between the PCB concentration in biota relative to the concentration sediments (Gobas and Morrison 2000). The BSAF is the ratio between the concentration of PCBs in biota to the concentrations of PCBs in sediment and can be used to predict the concentrations in biota (Alava et al. 2012b). The BSAF can be used to understand whether the contaminant is likely to biomagnify in food webs (Desforges et al. 2014).

Alava et al. 2012 reported the BSAF for Chinook Salmon and Resident Killer Whales for seven regions where the whales spend time throughout the year (Alava et al. 2012b). The reported BSAF values were theoretical as they assumed 100% of time was spent by SRKW or Chinook salmon in each region. Whereas in reality, Chinook salmon and SRKW only spend a portion of the time in each region. Therefore, the BSAF values ($BSAF_j$) reported by Alava et al. 2012 were multiplied by the annual proportion of time spent ($\emptyset_j$) by SRKW in the Salish Sea region (63%) and the proportion of time spent by Fraser River Chinook salmon in the Salish Sea (28.84%) (Equation 2). The log BSAF in Chinook salmon ($BSAF_{SS}$) was 1.36 $\pm$ 0.18 kg/kg dry weight in the Salish Sea. The log BSAF for all adult male SRKW ($BSAF_{KW}$) was 4.30 $\pm$ 0.33 kg/kg dry weight and the log BSAF for adult female SRKW ($BSAF_{KW}$) was 3.35 $\pm$ 0.33 kg/kg dry weight. These BSAF values for Chinook salmon and SRKW in the Salish Sea region were used with current concentrations of PCBs in sediments (C_s) to estimate the concentrations of ∑PCB in Chinook salmon(C_{SS}) and killer whales (C_{KW}) (Equations 3 and 4) (Alava et al. 2012a).

The current concentration of PCBs in sediment (C_s) was represented by the geometric mean concentration of ∑PCB in sediments in the Salish Sea (Strait of Georgia, SRKW Critical Habitat in Canada, and SRKW Critical Habitat in Juan de Fuca Strait and Puget Sound) from 2010 – 2017. The mean was log 10 transformed for the BSAF calculations (Equation 3 and 4). The time period of 2010 to 2017 was used for this analysis because previous PCB food web modelling for Resident Killer Whales used sediment PCB concentration data for the Salish Sea from 2002 to 2009 (Lachmuth et al. 2010; Alava et al. 2012a).

The logarithmic concentration of PCBs in Chinook salmon ($\log C_{SS}$) was estimated by adding the log BSAF for Chinook salmon ($\log BSAF_{SS}$) to the logarithm of the observed total concentrations of PCBs sediment ($\log C_s$) (Equation 3). The logarithmic concentration of PCBs in SRKW ($\log C_{kw}$) was estimated by adding the log BSAF for killer whales ($\log BSAF_{KW}$) to the logarithm of the observed total Concentrations of PCBs in sediment ($\log C_s$) (Equation 4). The uncertainty (*SD*) in the model inputs (i.e. log BSAF) and model calculations (i.e. log BSAF) were propagated in the estimate of the concentration of PCBs in biota (C_B) (Equation 5). The model estimates of total concentrations of PCBs in Chinook salmon and SRKW were compared to the observed total concentrations of PCBs in Chinook salmon and SRKW.

$$BSAF = BSAF_j \times \emptyset_j \tag{2}$$

$$\log C_{SS} = \log C_s + \log BSAF_{SS} \tag{3}$$

$$\log C_{KW} = \log C_s + \log BSAF_{KW} \tag{4}$$

$$SD_{CB} = \sqrt{(SD_{CS}{}^2 + SD_{BSAF}{}^2)} \tag{5}$$

Model performance

To assess the model performance, the overall model bias (MB) was calculated for Chinook salmon, adult male and adult female SRKW (*i* = 1 to 3). The model bias provides a way to quantitatively compare the model estimated and the observed total Concentrations of PCBs (Equation 6) (Alava et al. 2012a). A model bias of 1 indicates the model estimates on average match the observed data. A model bias of > 1 indicates the model is systematically over – estimating and a model bias of < 1 indicates the model is systematically under - estimating (Arnot and Gobas 2004). The standard deviation of the model bias was also calculated.

$$MB = 10^{\left(\sum_{i=1}^{n} \frac{[log\ (Estimated\ C_{kw_i} / Observed\ C_{kw_i})}{n}\right)} \tag{6}$$

3.2.3. Half-life of PCBs

Food web bioaccumulation model description

The half - life of PCB congeners in Chinook salmon and SRKW were estimated using a previously developed food web bioaccumulation model built in Excel (Lachmuth et al. 2010; Alava et al. 2012a, 2016). The food web bioaccumulation model was designed for the resident killer whale life cycle history and food chain (Figure 2.2). The model structure and results of sensitivity analyses are documented elsewhere (Lachmuth et al. 2010; Alava et al. 2012b, 2012a, 2016). The food web bioaccumulation model uses inputs of environmental concentrations of PCBs in water and sediment and information on processes that control concentrations of PCBs in environmental media and biota (Lachmuth et al. 2010). An updated version of the model that includes additional prey species (e.g. other Pacific salmon) was used in this study (Excel file provided by Dr. Juan José Alava, *personal communication*, 2018). The model output of elimination rates was used to calculate the half – lives of individual PCB congeners in Chinook salmon and SRKW and an average half – life for ∑PCB.

Model inputs

Chinook salmon weight

The half – lives of PCB congeners in Chinook salmon were estimated for adult Chinook salmon weighing 10 kg. This weight was used because SRKW are reported to selectively prey on 4 to 5 year old Chinook salmon that are 8 to 13 kg (Ford and Ellis 2005).

Concentrations of individual PCB congeners in sediment

The main input to the food web bioaccumulation model was the concentration of individual PCB congeners in sediment. The geometric mean concentration of individual PCB congeners in sediment was calculated for the Strait of Georgia in BC from sediment samples collected from 2010 to 2017. The geometric mean concentrations of individual PCB congeners in sediment were calculated for Juan de Fuca Strait and Puget Sound in Washington from sediment samples collected from 2010 to 2016. In the Strait of Georgia, sediment PCB concentration data were collected for 34 out of the 40 PCB congeners included in the food web bioaccumulation model. In Washington, the concentrations of PCB congeners in sediment were collected for only 16 of the PCB congeners included in the model.

Environmental parameters

Several of the key environmental input parameters used in the model were updated for this study as recommended (personal communication Juan José Alava, 2018). These parameters included total organic carbon, water temperature, salinity, pH and dissolved oxygen. The average total organic carbon content was calculated as described for BC (southern Strait of Georgia including BC SRKW Critical Habitat) and Washington (Puget Sound and Juan de Fuca Strait). The other environmental parameters were obtained from publicly available databases. For BC, the values of the environmental parameters were obtained from the time series data from the Oceans Network Central Strait of Georgia station (Oceans Networks Canada 2018). The daily averages for water temperature, salinity and dissolved oxygen from 2010 to 2017 were exported from the Oceans Network website and the overall mean values were calculated in R. The pH in water in the Strait of Georgia was obtained from a DFO Canadian Science Advisory Secretariat (CSAS) report (Irvine and Crawford 2013). For Washington, the values of the environmental parameters were obtained from the Puget Sound long term water quality monitoring online database (Department of Ecology Washington State 2016). Since there are numerous stations in Puget Sound, the Main Basin station was selected to represent the region. The overall mean water temperature, salinity, pH and dissolved oxygen were exported from the water quality database for 2010 to 2016. A subset of the observations for deeper water were selected because they represented the conditions closer to the sediment on the seafloor. The overall mean values were calculated in R.

Half – life of PCBs in Chinook salmon and killer whales

The food web bioaccumulation model was used to estimate the half - life of PCBs in Chinook salmon and SRKW. The concentration of individual PCB congeners for BC and Washington were entered into the Excel version of the model (provided by Dr. J. J. Alava, 2018). The areas were kept separate because of the different composition of congeners. The model generates estimates of the uptake and elimination rates of each PCB congener. The overall elimination rate (k_T) in Chinook salmon was calculated by summing the fecal egestion (k_E), gill elimination (k_2) and growth dilution (k_G) elimination rates (Equation 7). The metabolic biotransformation of PCBs was assumed to be negligible for Chinook salmon as was previously reported (Alava et al. 2012a). The overall elimination rate (k_T) of PCBs in adult male, adult female and juvenile SRKWs were calculated by summing the rate constants for fecal egestion (k_E), urine excretion (k_U), lung elimination (k_{D}), growth dilution (k_G), metabolic transformation (k_M), lactation rate (k_L) and reproduction (k_R), where applicable (Equation 8). The half – lives for

Chinook salmon and killer whales were calculated by dividing the natural log of 2 (0.693) by the sum of the elimination rates (k_T) (Equation 9).

$$Chinook\ salmon\ k_T = k_E + k_2 + k_G \tag{7}$$

$$Killer\ Whale\ k_T = k_E + k_{\mathrm{U}} + k_D + k_M + k_{\mathrm{L}} + k_R \tag{8}$$

$$t_{1/2} = \frac{0.693}{k_T} \tag{9}$$

3.3. Question 3: Does the composition of PCB congeners in sediment, Chinook salmon and SRKW provide clues on sources of PCBs to SRKW?

3.3.1. PCB congener patterns in sediment

The fraction of each PCB congener was calculated for all sediment samples collected in BC (including both the Strait of Georgia and BC SRKW Critical Habitat) and in Washington (including both US SRKW Critical Habitat in Juan de Fuca Strait and Puget Sound). In the BC dataset, the concentration of individual PCB congeners in sediment (C_s) was reported for all surveys conducted from 2010 to 2017. Therefore, to be consistent, the same time period was selected from the Washington dataset. The concentration of individual PCB congeners and the ∑PCB concentration in each sample was calculated. The relative contribution of individual PCB congeners in each sediment sample was calculated by dividing the concentration of each PCB congener by the ∑PCB concentration in each sample (Equation 10). The average contribution of each PCB congener and standard deviation were calculated for all of BC (Strait of Georgia and BC SRKW Critical Habitat) sediment samples and all Washington (US SRKW Critical Habitat in Juan de Fuca Strait and Puget Sound) sediment samples (Equation 11). BC and Washington datasets were kept separate because of the differences in PCB congeners and sampling protocols. Only individual PCB congeners were used in these calculations, thus co - eluting PCB congeners or Aroclor mixtures were not included.

$$\%\ PCB\ congener_{sample} = \frac{Sample\ PCB\ congener\ C_s}{Sample\ \sum PCB_{C_s}} \times 100\% \tag{10}$$

$$Relative\ contribution\ of\ each\ PCB\ congener = \frac{\sum_{n}^{i=1}\ \%\ PCB\ congener_{sample}}{N_{Sample}} \tag{11}$$

3.3.2. PCB congener patterns in Chinook salmon

The PCB concentration data for the Chinook salmon collected in 2014 were used because individual PCB congener concentrations were available (Guy 2018). The percent contribution of each PCB congener was calculated by dividing the PCB congener concentration in each sample by the ∑PCB concentration in each Chinook salmon sample. Only individual PCB congener concentrations were used in this analysis, not co - eluting PCB congeners. The average contribution of each PCB congener and standard deviation were calculated for all Chinook salmon samples.

3.3.3. PCB congener patterns in SRKW

The measured concentrations of PCB congeners in the SRKW samples collected in 2015 were used in the PCB congener fraction analyses because individual congener concentrations were readily available in Jayda Guy's master's thesis (Guy 2018). For adult females (n = 6), the percent of each PCB congener was calculated by dividing the PCB congener concentration in each sample by the ∑PCB concentration in each sample. The average contribution of each PCB congener and standard deviation were calculated for female SRKW. Since there was only one adult male SRKW sample (age > 10 years old), the PCB congener fraction calculation for males is only based on one sample. Only individual PCB congener concentrations were used in this analysis, not co - eluting PCB congeners or Aroclor mixtures.

Chapter 4.

Results and Discussion

4.1. Question 1: What are the concentrations of PCBs in sediments of critical killer whale habitats and how do they differ spatially and temporally?

4.1.1. Spatial Trends in PCBs in Sediment

In the time period from 2010 to 2017, sediment samples were collected at sampling stations throughout the Strait of Georgia, Juan de Fuca Strait and Puget Sound (Figure 4.1). The map includes 812 data points representing individual sample points, with 540 of these samples were collected in BC Strait of Georgia (including SRKW Critical Habitat in Canada) and 272 of these samples were collected in the US SRKW Critical Habitat (Juan de Fuca and Puget Sound). The geometric mean ∑PCB concentration in sediment samples collected in the BC Strait of Georgia (including SRKW Critical Habitat in Canada) from 2010 to 2017 was 1,616 pg/g dry weight (upper *SD* = 5,054 pg/g dw; and, lower *SD* = 517 pg/g dw). The concentration of ∑PCB in sediment ranged from 85 pg/g to 210,130 pg/g in one sample. As stated in the methods section, the spatial analysis does not include the samples collected in 2015 and 2016 by Pollution Tracker program because coordinates were not published online for these samples. The geometric mean ∑PCB concentration in sediment samples collected in the US SRKW Critical Habitat from 2010 to 2016 was 24,945 pg/g dry weight (upper *SD* = 31,391 pg/g dw; and, lower *SD* =19,823 pg/g dw). While the geometric mean concentration of ∑PCB measured in sediment in the BC Strait of Georgia (including SRKW Critical Habitat in Canada) was below the CCME interim SQG of 21,500 pg/g dry weight (21.5 μg/kg dw) for marine sediments (CCME 2001), the geometric mean concentration of ∑PCB in sediments in the US SRKW Critical Habitat (Juan de Fuca and Puget Sound) is above the CCME interim SQG. However, the SQG have been found to not be protective of upper trophic level species, like killer whales (Alava et al. 2012a; Alava et al. 2016; Arblaster et al. 2015). The recommended SQG is recommended to be between 20 to 200 pg/g dry weight (0.02 to 0.2 μg/kg dw) depending on the toxicity reference concentration selected (Alava et al. 2012a; Arblaster et al. 2015). All concentrations of ∑PCB measured in sediment samples throughout the Salish Sea between 2010 and 2017 were above the lower recommended SQG of 20 pg/g dry weight and the majority (95%) of the ∑PCB in these samples were above 200 pg/g dry weight.

The map of all the spatially interpolated ∑PCB concentration estimates shows that there is variation in the ∑PCB concentration in sediment within the Salish Sea (Figure 4.1). The spatially averaged concentration of ∑PCB was elevated in Puget Sound (i.e. as high as 60000 pg/g dw) compared to the Strait of Georgia, i.e. as low as 60 pg/g dw, as shown in Figure 4.1. The concentrations of PCBs in sediment could vary spatially due to different levels of historic PCB contamination, local sources and localized environmental and biogeochemical conditions that affect processes of burial and volatization. Contaminants may be contained within Puget Sound due restricted ocean current circulation and movement of water between fjord-like areas and the Pacific Ocean (West et al. 2017). The concentration of contaminants in surface sediment depends on the local rates of sediment accumulation and mixing (Johannessen et al. 2008b). By the mouth of the Fraser River, the concentrations of PCBs would be expected to be lower than other areas in the Strait of Georgia because the sediment accumulation rate is higher in this area because of the sedimentary (alluvial) inputs from the Fraser River Watershed depositing sediments in the coastal marine environment and also bringing sediment into the ocean (Johannessen et al. 2008b). The benthic invertebrate community in any area will affect how much the sediment is mixed through bioturbation. In the Strait of Georgia where there is an active benthic community, mixing of sediments can serve to move the more contaminated sediment found in lower layers of sediment up to the surface (Johannessen et al. 2008b). Due to the spatial variability of concentrations of PCBs in sediment in the Salish Sea, marine organisms are exposed to different levels of PCB contamination depending on their area of residence. For Chinook salmon populations in Puget Sound, it has been shown that PCB concentration in their tissues varies depending on the area of residence (Missildine et al. 2005; O'Neill and West 2009). Consequently, the accumulation of PCBs by SRKW from their food web will depend on their prey's main habitat.

The spatial interpolation approach has several limitations that should be considered when viewing the map results (Figure 4.1). The IDW method assumes that there is an inverse relationship between values and distance (Fisher and Getis 2010). When applied to concentrations of PCBs in sediment, it is assumed that the concentrations of PCBs in sediment in nearby locations are more similar than the sediment concentrations of PCBs at locations further away. However, this is a basic method of predicting concentrations of contaminants in marine sediment when in the marine environment there are many biological, geographical and physical processes that can affect the concentration of contaminants. Interpolation does not account for any geographical features that prevent contaminant storage and transfer or physical processes in the marine environment such as sediment dispersion with ocean currents. Also the IDW interpolation technique is constrained because it will not estimate values that are above or below the observed maximum and minimum values at the observed sample points (Sutton et al. 2009). For instance, there could be areas with lower concentrations of PCBs in sediment, but the interpolation results are based on the concentration values in sediment at nearby sample points.

The sampling locations may represent areas known to be “hotspots” with high PCB contamination which could lead to over – estimates of the concentrations of PCBs in the areas that were not sampled. For example, sediment samples have been collected in harbours and at disposal at sea sites. This could be one reason that the interpolated averaged concentration of ∑PCB in sediment are higher than the geometric mean concentration for the Salish Sea. The spatial analysis is limited by the number of samples collected in each geographic region and the distribution of sampling locations within each region. The IDW interpolation result will be best when the data points are evenly distributed (Sutton et al. 2009), which is unrealistic for marine sediment surveys. There were also gaps in the spatial coverage of sediment surveys within the SRKW Critical Habitat, particularly in Juan de Fuca Strait (Figure 4.1).

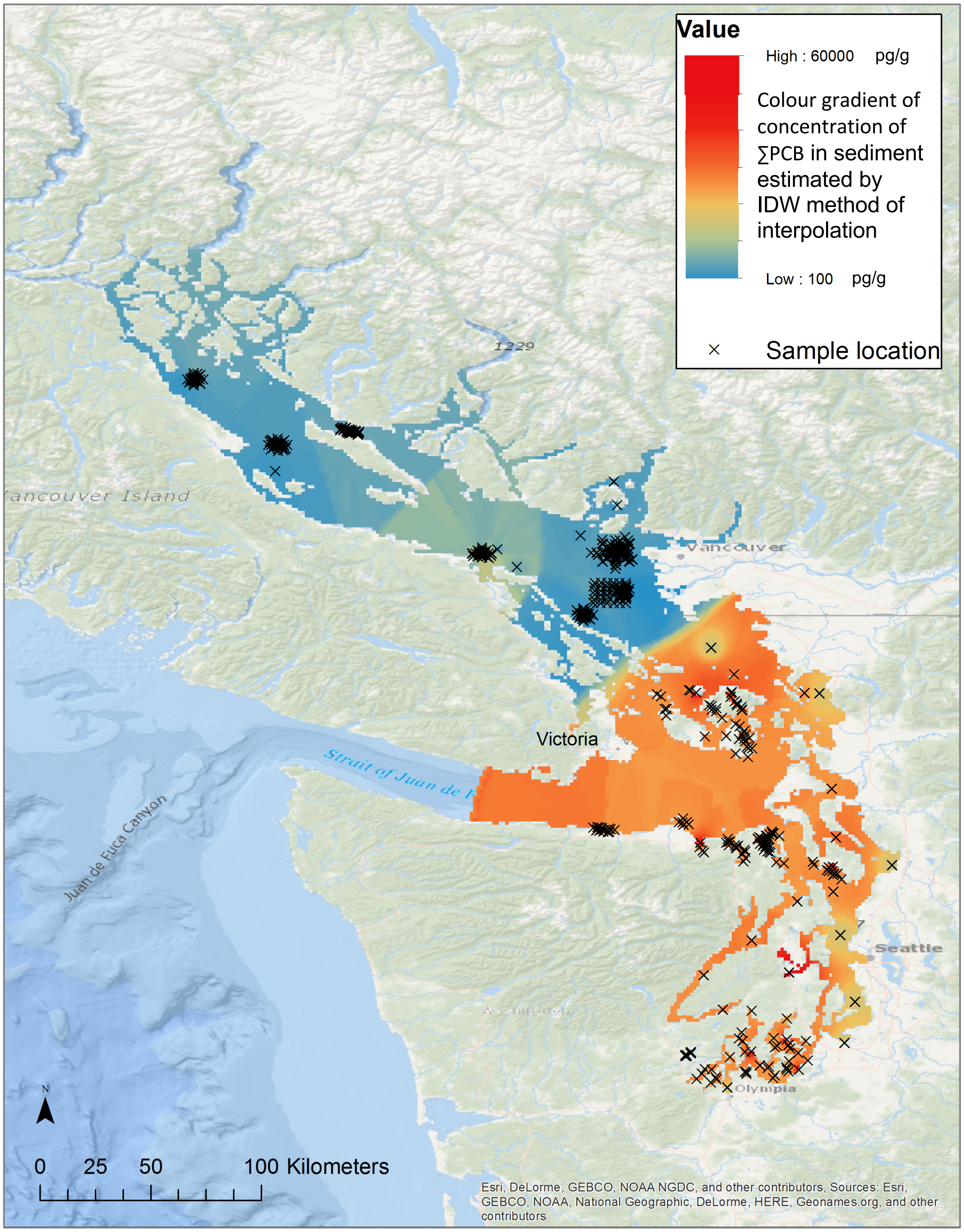

Figure 4.1. **Spatially interpolated log 10 transformed concentrations of ∑PCB in sediment (dry weight) in the Salish Sea. Interpolation was performed in ArcMap using the Inverse Distance Weighting (IDW) method.**

Source: Map created in ESRI software using Oceans base map (ESRI 2016a).

4.1.2. Temporal Trends of PCBs in Sediment

British Columbia Strait of Georgia and SRKW Critical Habitat

The concentrations of ∑PCB in the southern Strait of Georgia and BC SRKW Critical Habitat) varied greatly within years (Figure 4.2 and Table 4.1). In BC (Strait of Georgia and BC SRKW Critical Habitat), the geometric concentration of ∑PCB in sediment was highest in the 2013 samples (2859 pg/g dw) and the lowest geometric mean ∑PCB concentration was observed in the 2014 samples (152 pg/g dw) (Figure 4.2 and Table 4.1).The ANOVA indicated that there was a significant difference in at least one of the log 10 transformed mean concentrations of ∑PCB in sediment measured in samples collected between 2002 and 2017. A further year to year comparison using the Tukey HSD method showed that most of significantly different results occurred in comparisons to the low concentration of ∑PCB in sediment in the 2014 samples (Appendix B and C). According Tukey HSD results, there was not a statistically significant difference between the log 10 transformed mean concentration of PCBs in BC sediment in 2002 and the log 10 transformed concentration in 2017 (p – value = 1) (Appendix B and C).

Simple linear regression of the concentrations of ∑PCB in sediment measured in the BC Strait of Georgia by year produced a slope of -0.04 per year on the natural log scale with an associated p – value of 0.3. This suggests a not significant decline of 4% per year in the concentration of PCBs in sediment in the Strait of Georgia. The R2 value was 0.008 indicating that a very low proportion of the total variation of total concentrations of PCBs in sediment in BC was captured by linear regression by year (Figure 4.2) (Fox 1997). This result should also be interpreted with caution because of the unbalanced sampling design. The simple linear regression of the geometric mean concentrations for each year did not show a significant decrease in concentration of PCBs in sediment (slope -0.0041, p – value = 0.94) (Figure 4.3). The R2 value for the linear regression of the geometric mean concentrations of PCBs in sediment in the Strait of Georgia was 0.0007 (Figure 4.3).

Together, the results of the linear regression and the Tukey HSD test suggest that there has not been a substantial decrease in the ∑PCB concentration in sediments in the Strait of Georgia in the last 15 years. Given the half – life of PCBs in surface sediments in the Strait of Georgia is approximately 10 years (+/ - 8 years) (Lachmuth et al. 2010), a decline in the observed concentrations of PCBs in sediment would be expected in this time period if there were no new inputs of PCBs into the local marine environment. The concentration of PCBs could be expected to decline by half in 2 to 20 years in the absence of any new sources of PCBs into the Strait of Georgia marine environment.

Washington SRKW Critical Habitat

In Washington (US SRKW Critical Habitat in Juan de Fuca and Puget Sound), the concentrations of ∑PCB in sediment were greater than those measured in BC for all years (Table 4.2). The highest geometric mean PCB concentration in sediment was in 2005 samples (35,398 pg/g dw), while the lowest geometric mean concentration of ∑PCB was observed in the 2002 samples (11,578 pg/g dw) (Table 4.2). According to Tukey HSD test, there was a significant difference in the mean log 10 transformed concentrations of PCBs in sediment between 2016 and 2002 (p – value = <0.01) (Appendix B).

Simple linear regression of the concentrations of ∑PCB measured in sediment by year in US SRKW Critical Habitat produced a slope of 0.05 on the natural log scale with an associated p – value = <0.01. This indicates an increase of approximately 5% per year in the concentration of ∑PCB in sediment in US SRKW Critical Habitat. The R2 value was 0.24 indicating that approximately 20% of the variation in the concentration of ∑PCB in sediment in the US SRKW Critical Habitat was captured by linear regression by year (Figure 4.4) (Fox 1997). The simple linear regression of the geometric mean concentrations for each year also indicated a statistically significant increase of approximately 5% per year in the concentration of ∑PCB in sediment in US SRKW Critical Habitat (slope 0.05, p - value = 0.04) (Figure 4.5). The R2 value for the linear regression of the geometric mean concentrations was 0.31 (Figure 4.5). All three methods of investigating the change in concentrations of PCBs over time indicated there has been a significant increase in concentrations of PCBs in sediment overall in the US SRKW Critical Habitat in Puget Sound and Juan de Fuca based on the sampling results.

Given the half – life was estimated to be approximately 6.5 years (+/- 3.1 years), it would be expected that there would be a decline in the total concentrations of PCBs within central Puget Sound. Extrapolating beyond central Puget Sound, it would be reasonable to expect a decline in the total concentrations of PCBs in sediment in the US

SRKW Critical Habitat within the 14 year period from 2002 to 2016. The environmental conditions and inputs from potential local sources in other areas of Puget Sound and Juan de Fuca will lead to different sedimentation rates and PCB half – lives, but there will be some burial of PCBs by natural sedimentation and with no ongoing input in PCBs, then there would be some evident decline in concentrations of PCBs in sediment.

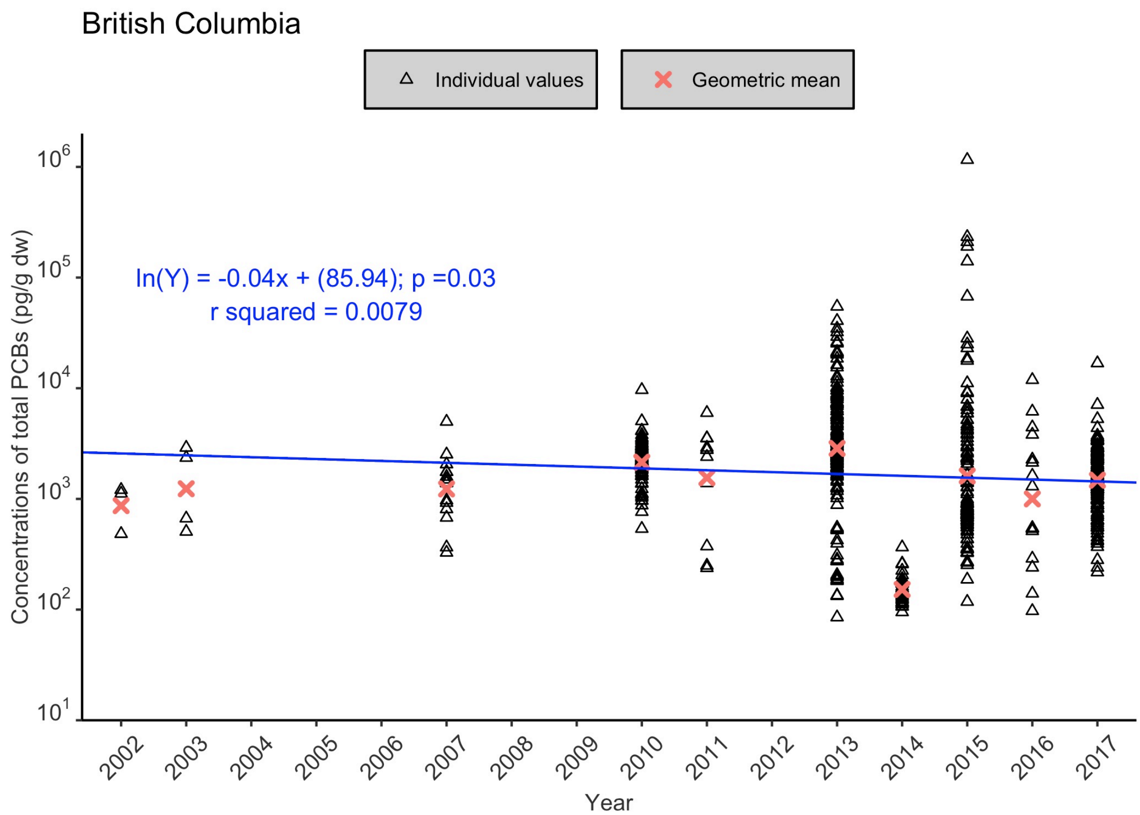

Figure 4.2. **Temporal trend showing the log 10 transformed concentrations of ∑PCB (pg/g dw) in sediment in British Columbia in the Strait of Georgia and SRKW Critical Habitat from 2002 to 2017. The linear regression line equation is shown on the natural log scale.**

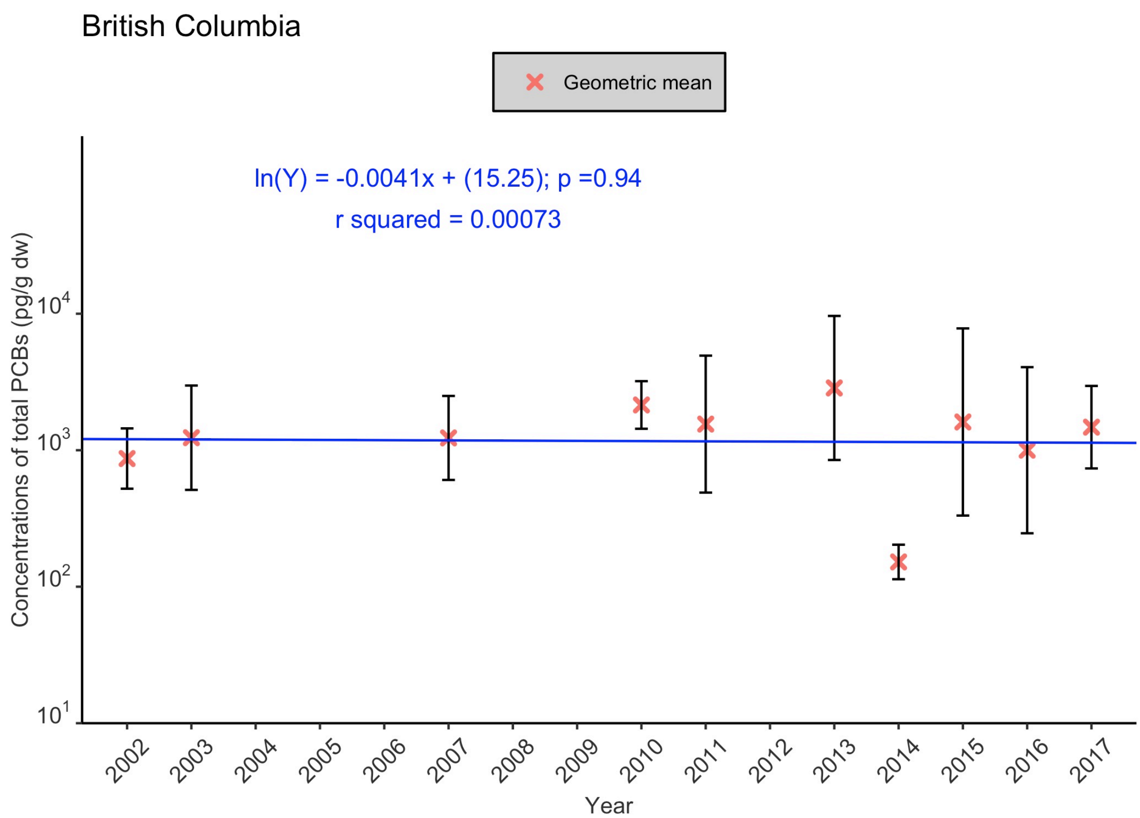

Figure 4.3. **Log 10 transformed average concentrations of ∑PCB (pg/g dw) in sediment in British Columbia in the Strait of Georgia and SRKW Critical Habitat from 2002 to 2017. The error bars indicate the standard deviation. The linear regression line equation is shown on the natural log scale.**

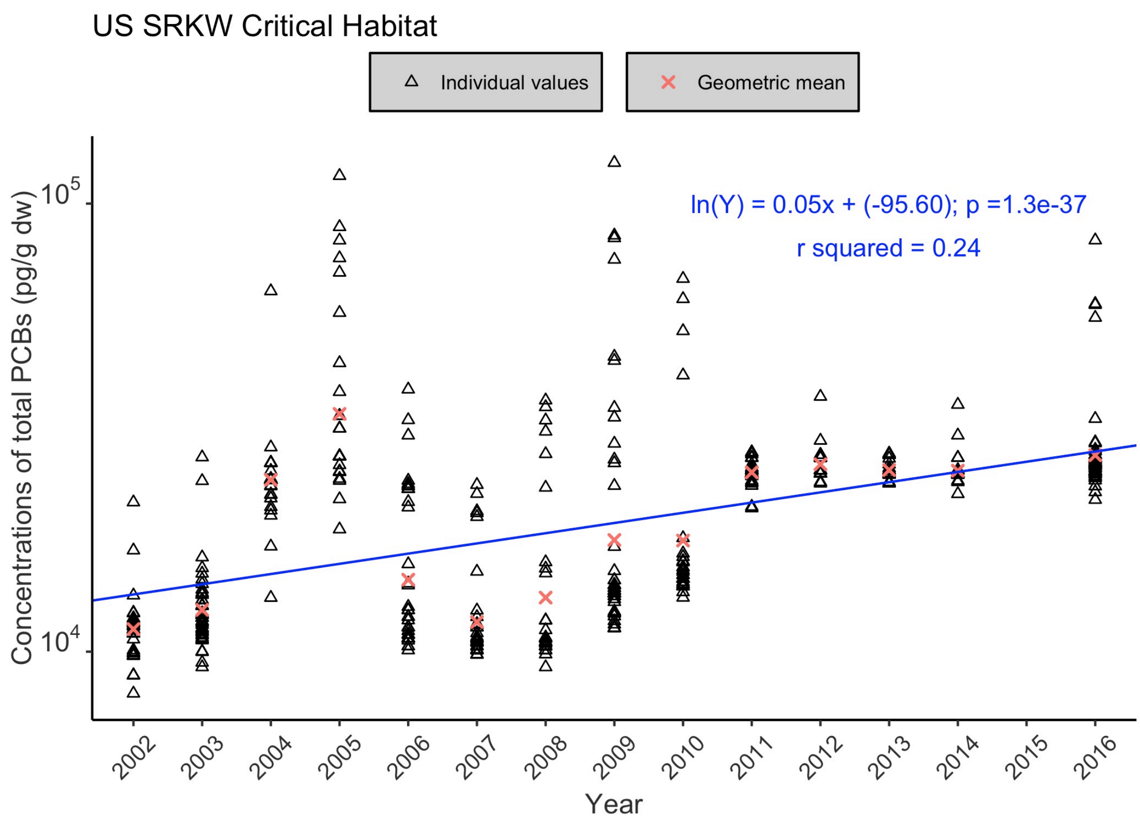

Figure 4.4. **Temporal trend showing the log 10 transformed concentrations of ∑PCB (pg/g dw) in sediment in Washington SRKW Critical Habitat from 2002 to 2016. The linear regression line equation is shown on the natural log scale.**

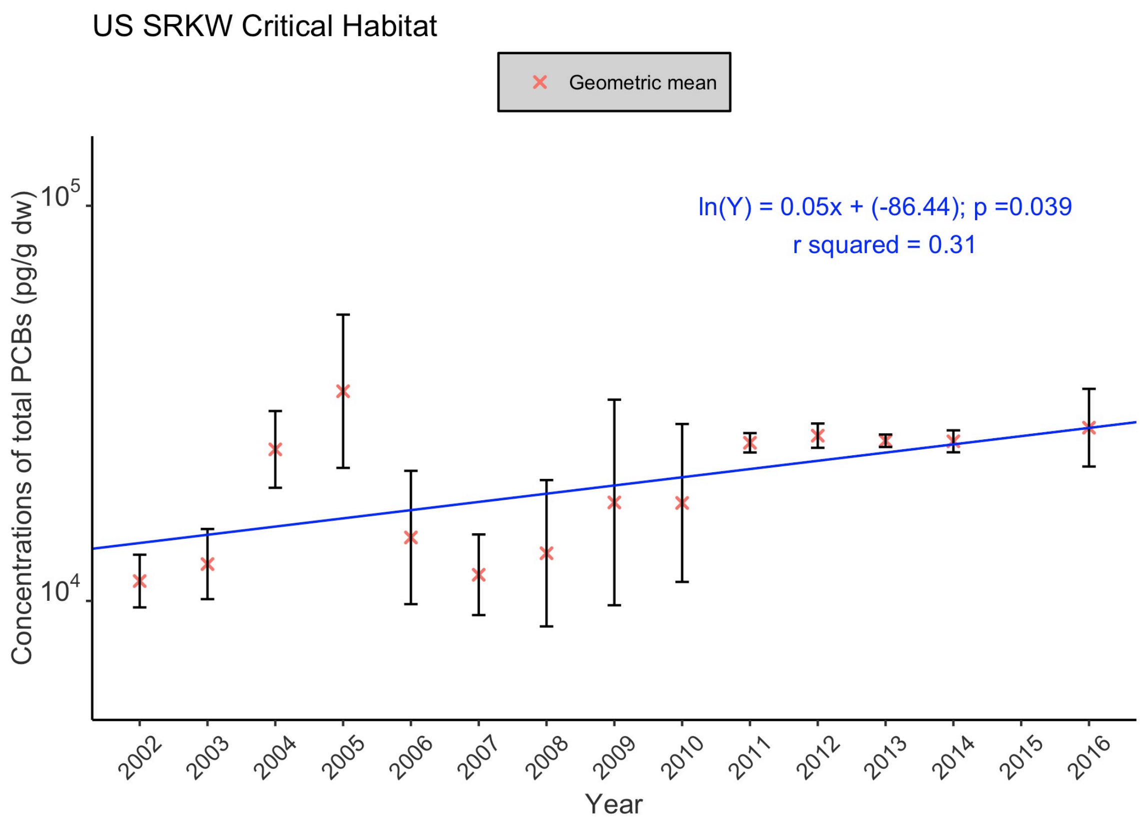

Figure 4.5. Log 10 transformed average concentrations of ∑PCB (pg/g dw) in sediment in Washington SRKW Critical Habitat from 2002 to 2016. The error bars indicate the standard deviation. The linear regression line equation is shown on the natural log scale.

Table 4.1. Summary of concentrations of ∑PCB (pg/g dw) in sediment samples collected in British Columbia in the Strait of Georgia and SRKW Critical Habitat from 1997 to 2017.

Year	Sample size	Geomean (pg/g dw)	Upper SD (pg/g dw)	Lower SD (pg/g dw)	Data Source
2002	3	869	1444	523	Johannessen et al. 2008, Wright et al. 2008, Grant et al. 2012
2003	4	1235	2978	512	Johannessen et al. 2008, Wright et al. 2008, Grant et al. 2012
2007	15	1230	2499	606	Johannessen et al. 2008, Wright et al. 2008, Grant et al. 2012
2010	101	2146	3205	1437	Ross et al . 2011 *
2011	11	1554	4933	489	Ross et al . 2012 *
2013	156	2859	9633	848	ECCC *
2014	34	152	203	113	ECCC *
2015	122	1611	7807	332	ECCC *, Pollution Tracker, 2018
2016	16	1000	4062	246	ECCC *, Pollution Tracker, 2018
2017	132	1475	2959	736	ECCC *

*Unpublished data from Environment and Climate Change Canada, Disposal at Sea Program provided in 2017.

Table 4.2. Summary of concentrations of ∑PCB (pg/g dw) in sediment samples collected in Washington SRKW Critical Habitat from 2002 to 2016.

Year	Sample Size	Geomean (pg/g dw)	Upper SD (pg/g dw)	Lower SD (pg/g dw)	Data Source
2002	43	11229	13092	9631	EIM database
2003	43	12397	15205	10107	EIM database
2004	33	24179	30239	19334	EIM database
2005	33	33932	53031	21711	EIM database
2006	43	14474	21342	9816	EIM database
2007	43	11648	14731	9210	EIM database
2008	33	13204	20221	8622	EIM database
2009	50	17752	32314	9752	EIM database
2010	30	17700	28039	11173	EIM database
2011	44	25133	26593	23753	EIM database
2012	43	26196	28116	24407	EIM database
2013	43	25428	26367	24522	EIM database
2014	46	25349	27022	23779	EIM database
2016	66	27441	34408	21885	EIM database

EIM database (Washington State Department of Ecology 2018a)

4.1.3. Temporal Trends of PCBs in Biota

Chinook salmon

The geometric mean and standard deviation of the concentration of ∑PCB in Chinook salmon samples were calculated for 2000 and 2014 (Table 4.3). Linear regression of the logarithm of the ∑PCB concentration in Chinook salmon showed there were not a statistically significant difference in the concentration of PCBs in Chinook salmon between 2000 and 2014 (Figure 4.6). This suggests that there hasn't been a decline in the concentration of ∑PCB in Chinook salmon. However, these results should be interpreted with caution because of the low sample sizes and only two monitoring years. Since the stock was not identified for the Chinook salmon samples, it is only assumed that the samples are comparable because both sets of Chinook samples were collected in BC waters. The stock and whether the fish are stream – type or ocean – type salmon are important for understanding the exposure history of the Chinook salmon over its lifespan and whether the fish are representative of typical prey of SRKW.

This study did not include Chinook salmon that have been collected in Puget Sound and analyzed for PCBs (O'Neill et al. 1998, 2006; O'Neill and West 2009; Hope 2012). This would be a good extension of the study because it would provide a better picture of the regional accumulation trends of PCBs in Chinook salmon throughout the Salish Sea.

Table 4.3. Geometric mean and standard deviations of concentrations of ∑PCB (mg/kg lw) in Chinook salmon samples collected in 2000 and 2014.

Year	Sample Size	Geomean	Upper SD	Lower SD	Data Source
2000	12	0.28	1.01	0.08	Cullon et al. 2009 *
2014	7	0.28	0.46	0.18	Guy et al. 2018

* As reported in Guy et al. 2018

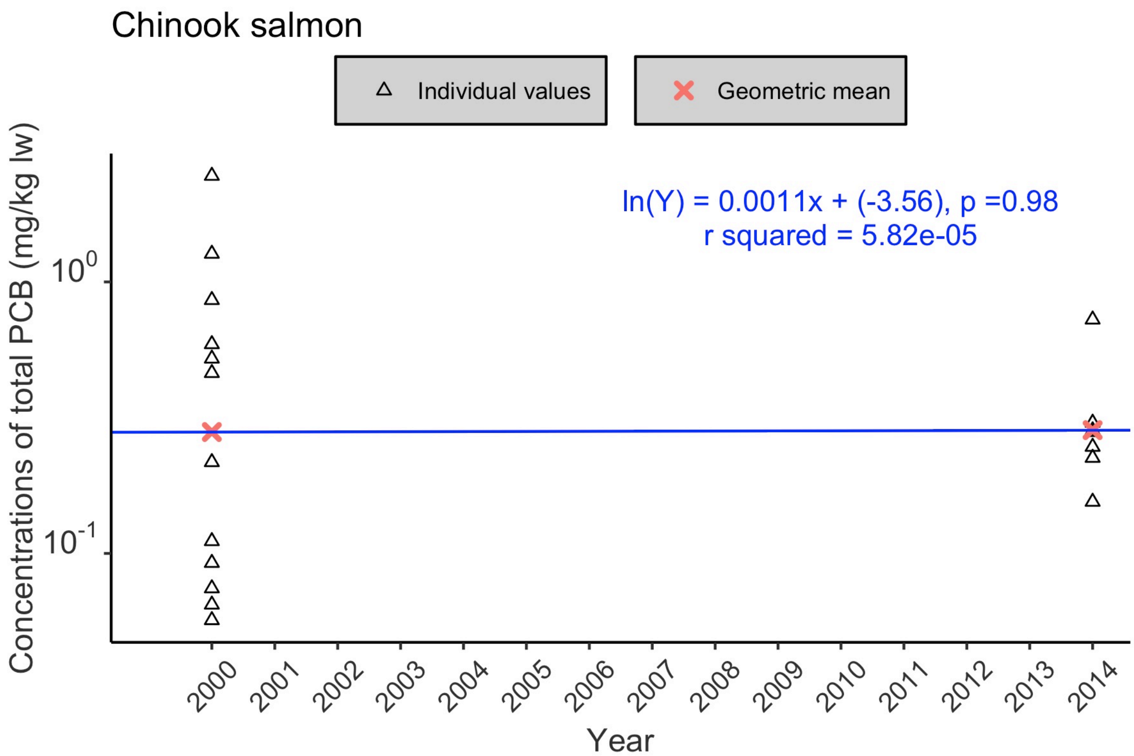

Figure 4.6. Temporal trend of log 10 transformed oncentrations of ∑PCB (mg/kg lipid weight) in Chinooks salmon samples collected in 2000 and 2014. The linear regression line equation is shown on the natural log scale.

Southern Resident Killer Whales

The annual geometric mean and standard deviation were calculated for all adult female SRKW samples collected between 1996 and 2015 (Table 4.4) and all adult male SRKW samples collected between 1993 and 2015 (8

Table 4.5). The Tukey HSD test results showed that there was no significant difference in the log 10 transformed mean ∑PCB concentration in adult females between any years (Appendix A).

Simple linear regression of the log transformed concentrations of ∑PCB in adult female SRKW shows a declining trend that is not statistically significant (*p – value* = 0.07) (Figure 4.7). For adult female SRKW, the estimated slope of the regression fit was -0.09 on the natural log scale. This suggests the concentration of PCBs in female SRKW decreases by 9% per year. Simple linear regression of the log transformed concentrations of ∑PCB in adult male SRKW showed there were not a statistically significant decline over the sample time period (*p – value* = 0.24) (Figure 4.7). For adult male SRKW, the estimated slope of the regression fit was -0.05 on the natural log scale. This suggests the concentration of PCBs in male SRKW decreases by 5% per year.

The analysis of Concentrations of PCBs in SRKW is limited by small sample sizes and the assumptions inherent in comparing and combining data collected over several decades. The small sample size and large variability in the concentrations of PCBs in many individuals makes it challenging to assess PCB concentration trends over time. When the samples are grouped by sex and stage the maximum sample size is 6 adult male or females and the minimum samples size of 1 male or female per sampling event. In the linear regression, data for one individual were included because of the limited data available. This temporal analysis of the mean ∑PCB concentration across all sampled SRKW does not incorporate individual history of PCB exposure and accumulation. This would be important to consider because the concentrations of PCBs accumulated by killer whales depends on several factors including dietary preference, calving order (Ross et al. 2000), high PCB contaminated sediments versus low PCB contaminated sediments in habitats (Alava et al. 2012a), reproductive history birth year and pod membership (Mongillo et al. 2012).

Table 4.4. Geometric mean and standard deviations of concentrations of ∑PCB in adult female SRKW samples collected from 1996 to 2015 in the Salish Sea.

Year	Geomean	Upper SD	Lower SD	Sample Size	Source
1996	50.9	87.6	29.6	2	Ross et al. 2013 *
2006	45.0	NA	NA	1	Krahn et al. 2007 *
2007	17.8	70.1	4.5	6	Krahn et al. 2009 *
2015	10.3	26.2	4.0	6	Guy et al. 2018

* As reported in Guy et al. 2018

Table 4.5. Geometric mean and standard deviations of concentrations of ∑PCB in adult male SRKW samples collected from 1993 to 2015 in the Salish Sea.

Year	Geomean	Upper SD	Lower SD	Sample Size	Source
1993	31.0	321.4	3.0	2	Ross et al. 2013 *
1996	110.2	241.7	50.2	2	Ross et al. 2013 *
2000	248.0	NA	NA	1	Ross et al. 2013 *
2004	22.2	39.8	12.4	6	Ross et al. 2013 * Krahn et al. 2007 *
2006	78.2	139.6	43.8	4	Krahn et al. 2009 *
2007	30.8	38.8	24.4	3	Krahn et al. 2009 *
2015	10.3	NA	NA	1	Guy et al. 2018

* As reported in Guy et al. 2018

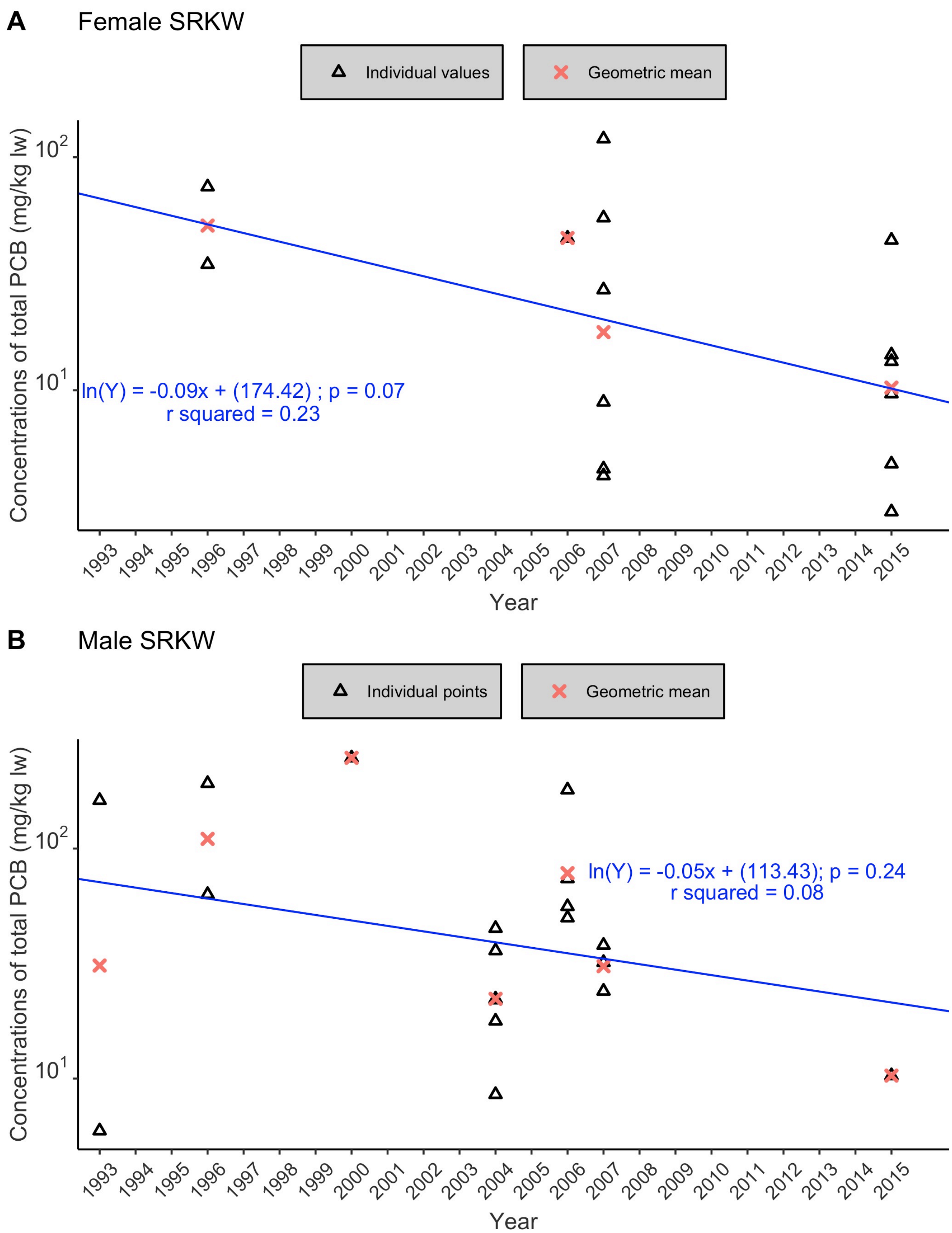

Figure 4.7. Temporal trends of log 10 tranformed concentrations and geometric mean of ∑PCB (mg/kg lipid weight) in SRKW including adult female (A) and male (B) samples collected from 1993 to 2015 in the Salish Sea. The linear regression line equation is shown on the natural log scale.

4.2. Question 2: Contribution of PCBs in Sediment to PCBs in SRKW food web. Can the concentrations of PCBs in the habitat of SRKWs contribute significantly to the concentration of PCBs observed in Chinook salmon and SRKW?

4.2.1. Comparison of concentrations of PCBs in sediment and biota

Bioaccumulation is illustrated in the plot of the observed concentrations of ∑PCB in sediment, Chinook salmon and SRKW (Figure 4.8). The geometric mean ∑PCB concentration in sediment in the Strait of Georgia and BC SRKW Critical Habitat was 0.11 mg/kg organic carbon (or 0.32 mg/kg equivalent lipid). The geometric mean concentration of ∑PCB in sediment in US SRKW Critical Habitat (Juan de Fuca and Puget Sound) was 1.63 mg/kg organic carbon (or 4.66 mg/ kg equivalent lipid). Once PCBs are taken up by biota at lower trophic levels, the concentration of PCBs would be expected to biomagnify in the food web of Chinook salmon. However, when the concentrations of PCBs in sediment are converted to lipid equivalent concentrations, there is not a significant difference between the concentration of ∑PCB in sediment in the BC Strait of Georgia and the lipid normalized concentration in the Chinook salmon samples collected in BC. The geometric mean concentration of ∑PCB in Chinook salmon samples collected in 2000 and 2014 was 0.28 mg/kg. As discussed previously, there are very limited data available on the concentration of PCBs in Chinook salmon, precluding a more concerted and statistically robust comparison and these values may not be representative of the concentration of PCBs in Chinook salmon in the SRKW food web. The study could be expanded to compare the concentration of PCBs in Chinook salmon originating from Puget Sound to the concentrations of PCBs in the relatively more contaminated sediment in US SRKW Critical Habitat. Differences in organic carbon content in sediments (i.e. 1.50%) from the Strait of Georgia versus that found in sediments (i.e. 1.58%) from Puget Sound in Washington can also influence the uptake and trophic transfer of PCBs in the food web (Alava et al 2012a).

The concentration of PCBs in SRKW would exceed the concentration in Chinook salmon due to trophic transfer and biomagnification of PCBs at each trophic level in the food web and the long life span of killer whales. As shown in Figure 4.8, the concentration of PCBs observed in SRKW is several magnitudes greater than the concentration of PCBs observed in Chinook salmon. The geometric mean ∑PCB concentration for adult female SRKW samples collected from 1996 to 2015 was 17.46 mg/kg lipid weight, which is approximately 62 times greater than the concentration of PCBs in Chinook salmon (Figure 4.8). The geometric mean ∑PCB concentration for adult male SRKW was 40.74 mg/kg lipid weight, which is approximately 145 times greater than the concentration of PCBs in Chinook salmon (Figure 4.8).

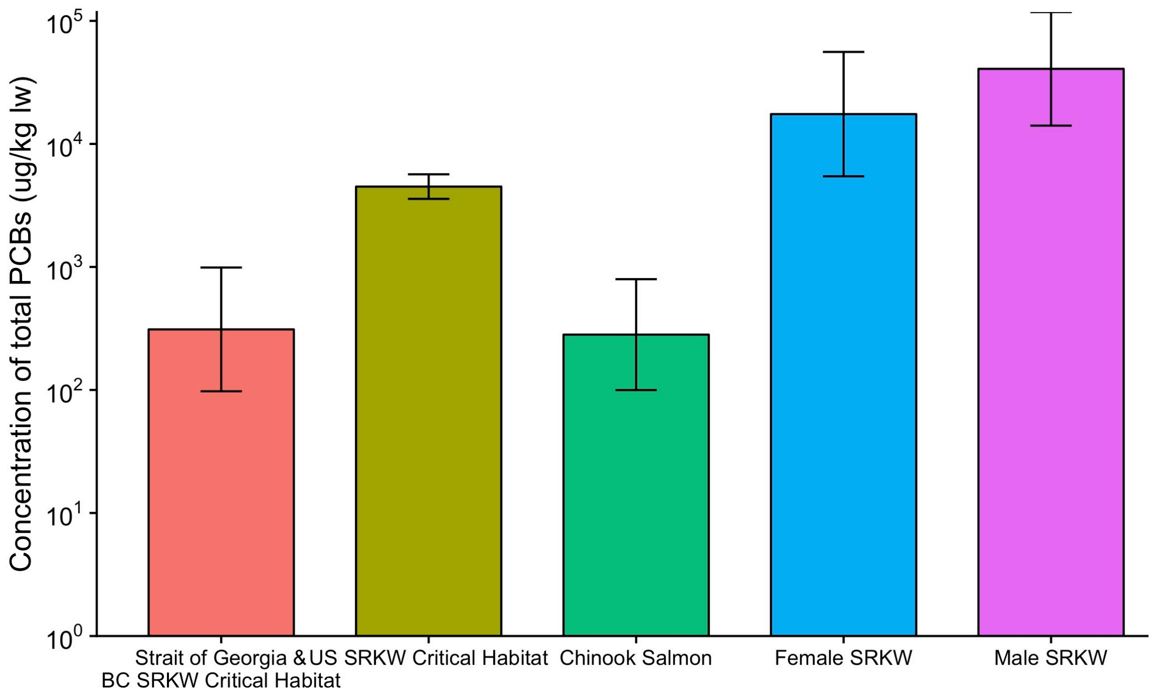

Figure 4.8. Comparision of the lipid normalized geometric ∑PCB concentration (ug/kg) in sediment, Fraser River Chinook Salmon and adult SRKW. All geometric means were calculated from data pooled from multiple years: sediment (2010 to 2017), Chinook salmon (2000 and 2014) and SRKW (1993 to 2015). The bars indicate the standard deviation.

4.2.2. BSAF estimated concentrations of PCBs in Chinook salmon and SRKW

The estimated concentrations of ∑PCB in Chinook salmon and SRKW were similar to the observed concentration of ∑PCB in Chinook salmon and SRKW (Figure 4.9). The estimated concentrations of PCBs in biota were within the standard deviation of the observed concentrations of PCBs in female SRKW, male SRKW and Chinook salmon (Figure 4.9). The overall model bias of 3.48 (upper standard deviation of 5.7, lower standard deviation of 2.15) indicated that on average the BSAF modelling approach overestimated the concentrations in biota. However, the standard deviation of the observed and estimated concentrations of PCBs in biota show that the estimated concentrations were not significantly greater than the observed concentrations (Figure 4.9). The BSAF model results indicate that the concentrations of PCBs in sediment in the Salish Sea have the potential to accumulate through the marine food web to the levels of PCBs observed in SRKW.

This approach assumes that the geometric mean concentration of ∑PCB in sediment samples collected from 2010 to 2017 in the Salish Sea accurately represents the concentrations of PCBs that SRKW are exposed to in this area through food web bioaccumulation. Individual PCB congeners will have different BSAF values based on the physiochemical properties of the congener (Desforges et al. 2014).

The exposure to PCBs originating from Salish Sea sediment depends on the time spent by both SRKW and their prey, particularly Chinook salmon in the Salish Sea. An effort was made to adjust for the time spent by SRKW and Chinook salmon in the Salish Sea past on known distributions reported by Alava et al. (2012a). However, this approach assumed that all three SRKW pods have the same annual distribution, but in reality the pods have a different distribution throughout the year (Alava et al. 2012a). There has been more recent research on the distribution of SRKW throughout the year and analysis of their key foraging areas which could lead to improved estimates of time spent in the Salish Sea region (Ford et al. 2017)

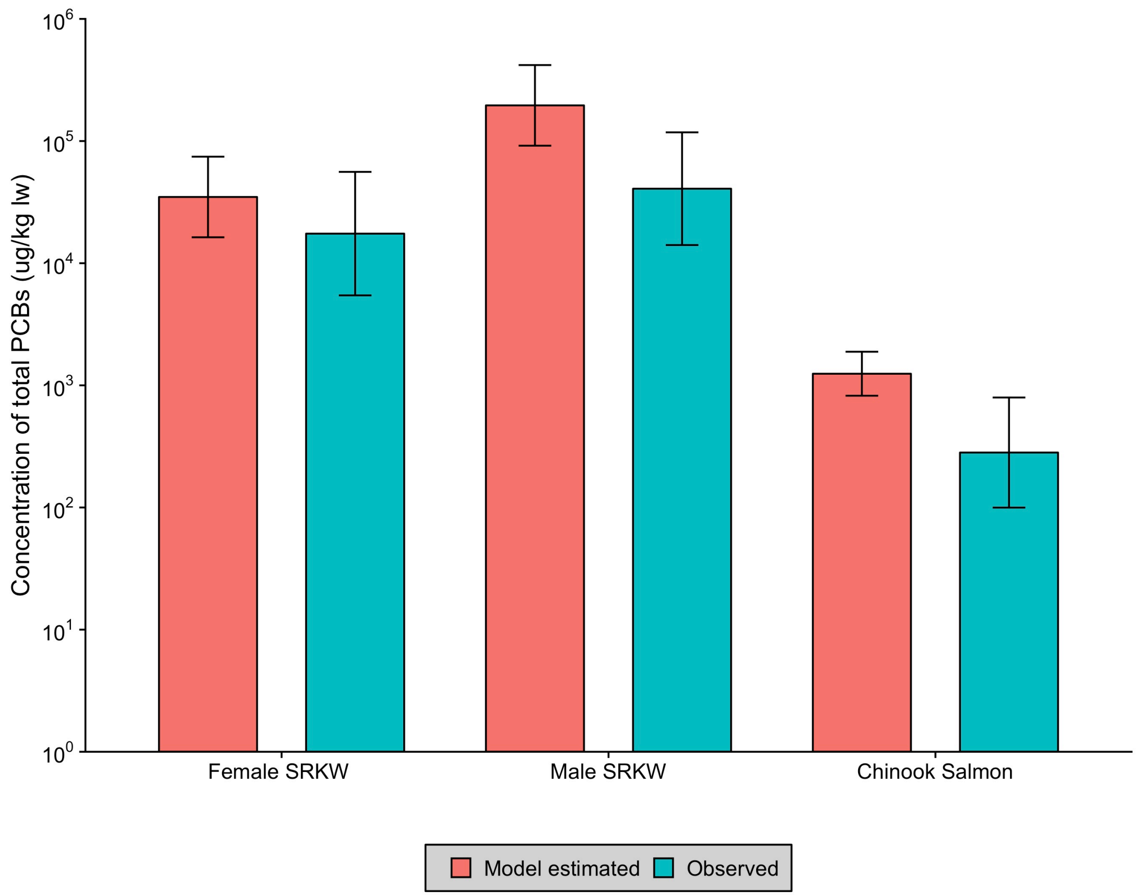

Figure 4.9. **Comparison of the model estimated total concentrations of PCBs in Chinook salmon and SRKW (ug/kg lw) to the geometric mean of the observed total concentrations of PCBs in Chinook salmon and SRKW (ug/kg lw). The error bars indicate the standard deviation.**

4.2.3. Half - life of PCBs in Chinook salmon and SRKW

Half – lives for PCB congeners in Chinook salmon were estimated based on the concentrations of PCB congeners in sediment in BC (Strait of Georgia and BC SRKW Critical Habitat) and Washington (SRKW Critical habitat in Juan de Fuca Strait and Puget Sound). The average half – life for all PCB congeners in Chinook salmon was approximately 1.05 years (Table 4.6). The length of the half - life depends on the PCB congener chemical properties. The estimated half- life in Chinook salmon was shortest (0.46 years) for PCB 8 (Table 4.6) which has a log K_{OW} of 5.27 (Alava et al. 2012b) . The longest estimated half – life in Chinook salmon was 1.68 years for PCB 194 (Table 4.6) which has a log K_{OW} of 8.12 (Alava et al. 2012b). The log K_{OW} can be used to describe the solubility in water and lipid and elimination rates increase with higher log K_{OW} (Borgå et al. 2004). Therefore, it makes sense that the shortest half - life occurs for the PCB congener with a lower log K_{OW} and the longest half – life was estimated for the PCB congener (PCB 194) with the longest half – life of the 39 PCB congeners include in the food web bioaccumulation model (Alava et al. 2012b).

Given that Chinook salmon spend 2 - 4 years of their life in the ocean, these half – lives confirm that PCBs would be accumulated in marine habitats rather than in freshwater as has been reported in previous research (O'Neill and West 2009). Whether PCBs are taken up by Chinook salmon in local coastal marine habitats or offshore will depend on the life history type ("stream" vs. "ocean") and migration route of the salmon. Chinook salmon that spend more time in residence in coastal waters (e.g. "ocean – type") of the Salish Sea could be expected to have concentrations of PCBs that reflect local concentrations of PCB congeners in sediment. The half – life of less than 0.5 years for some PCB congeners suggests that the half – life may be short enough for stream – type Chinook salmon to also be influenced by the local concentrations of PCBs in sediment when they return to coastal waters in the Salish Sea prior to spawning.

Half – lives for PCB congeners in adult and juvenile SRKW were estimated based on the concentrations of PCB congeners in sediment in the entire Salish Sea (Table 4.7). Adult male SRKW had the longest average PCB half – life of 11. 4 years. The average half – life in juveniles was 2.8 years and the average half – life in adult females was 1.6 years. The average half – life of PCB congeners in female SRKW was shorter than male SRKW because PCBs are eliminated through maternal transfer, including reproduction and lactation. The estimated half – lives were shortest for PCB 18 and longest for PCB 194 in juvenile, male and female SRKW. The log K_{OW} for PCB 18 was 5.62 (the second lowest log K_{OW} of all congeners included in the model) and the log K_{OW} for PCB 194 was 8.18 (Alava et al. 2012b). As discussed above, the time to eliminate an organochlorine contaminant is positively correlated with the log K_{OW} value (Borgå et al. 2004).

The estimated PCB half – lives in SRKW indicate that concentrations of PCBs in SRKW tissue could be expected to decline over the life time of these long – lived species if exposure to PCBs ceases. In adult males within 12 years, the concentrations would be expected to decrease by half. However, in the 22 year period from 1993 to 2015, the mean concentrations of PCBs in adult male SRKW did not exhibit a statistically significant decline. In adult females, a rapid decline in PCBs stored in their tissue would be expected to be observed within 2 years. These findings suggest that there is an ongoing source of PCBs into the SRKW marine food web.

Table 4.6. Summary of half - lives of PCB congeners in Chinook salmon estimated using the food web bioaccumulation model.

Region	Chinook salmon weight (kg)	Mean half - life (years)	Minimum half - life (years) (e.g. PCB 8)	Maximum half - life (years) (e.g. PCB 194)
BC Strait of Georgia and BC SRKW Critical Habitat	10	1.04	0.46	1.68
Washington SRKW Critical Habitat in Juan de Fuca and Puget Sound	10	1.05	0.61	1.66

Table 4.7. Summary of half - lives of PCB congeners in SRKW estimated using the food web bioaccumulation model.

Region	Stage and Gender	Mean half - life (years)	Minimum half - life (years) (e.g. PCB 8)	Maximum half - life (years) (e.g. PCB 194)
Salish Sea (Strait of Georgia and SRKW Critical Habitat in Canada and the US)	Adult male	11.40	8.88	11.75
	Adult female	1.59	1.51	1.60
	Juvenile	2.84	2.57	2.87

4.3. Question 3: Does the composition of PCB congeners in sediment, Chinook Salmon and SRKW provide clues on sources of PCBs to SRKW?

4.3.1. PCB congener patterns in sediment

The current study results can be used to identify the composition pattern of PCBs and the dominant PCB congeners in the sediment, Chinook salmon and SRKW samples. The most dominant PCB congeners in the BC Strait of Georgia sediment was PCB 118 followed by PCB 153, PCB 110, PCB 101, respectively (Figure 4.11). The PCB congeners with the highest fraction of ∑PCB in the Washington (Juan de Fuca and Puget Sound) sediment samples were PCB 128 and PCB195, followed by PCB 169 and PCB 209 (Figure 4.10). The findings may indicate that the PCBs released into each area have a different composition. However, this is difficult to conclude because only 21 PCB congeners were analyzed in the Washington sediment samples. Due to the limited congener data available, a more detailed comparison between PCB congener patterns in the sediment in Washington and BC could not be completed. To further investigate the differences in PCB congener patterns in sediment in different geographical regions, a potential next step is to designate smaller regions of interest and analyze the PCB congener concentration for each of the smaller regions.

A previous investigation of the pattern of PCB congeners in sediment in British Columbia from Hecate Strait to the Strait of Georgia found the dominant PCB congeners were 138, 153, 118, 101, 110 and 149 (Desforges et al. 2014). Notably, the current study did not detect PCB 138 as a dominant congener. This is likely because PCB 138 often occurred co-eluting groups of PCB congeners in the BC sediment data set and the congener pattern analysis was only completed for individually detected PCB congeners. This approach was used to avoid introducing false patterns by assigning a dominant congener to a group of PCB congeners.

4.3.2. PCB congener patterns in Chinook salmon and SRKW

The PCB congener contributing the most to the concentration of ∑PCB in the Chinook salmon samples was PCB 153 followed by PCB 129 (Figure 4.11). The PCB congener most to the concentration of ∑PCB in the male and female SRKW samples was PCB 153, followed by PCB 129 (Figure 4.11). The high contribution of PCB 153 in Chinook salmon is consistent with an earlier study by Cullon et al. (2012) that identified PCB 153 as a dominant congener in marine species at all trophic levels in the harbour seal diet in the Strait of Georgia (Cullon et al. 2012). PCB 153 has also been identified as one of the dominant PCB congeners in killer whales and other cetaceans (Tanabe et al. 1988; Ross et al. 2000). However, contrary to the current study, PCB 138 was also found to be a dominant congener in killer whales and other cetaceans studied, previously (Ross et al. 2000).

Several congeners, PCB 83, PCB 153, PCB 180, and PCB 187, are present in BC sediment, Chinook salmon and SRKW (Figure 4.11). This provides some indication that local PCB contaminated sediment influences the composition of PCB congeners observed in Chinook salmon or SRKW. However, the PCB congeners found to contribute the most to the concentration of PCBs in Chinook salmon and SRKW (PCB 153 and PCB 129) differ from the dominant congener profile observed in sediment in the BC and the US SRKW Critical Habitat. The PCB pattern in sediments is comprised by a higher abundance of low chlorinated PCB congeners relative to the PCB profile observed in Chinook salmon and SRKW, as shown in Figure 4.11A. This could be attributed to other factors that are recognized to change the composition of PCBs in different media, such as metabolism, biomagnification and transfer to other environmental media. This study also does not account for differences in PCB congener fractions observed in biota compared to sediment due to differing rates of bioaccumulation and metabolism of individual PCB congeners. The PCB congeners with more chlorine atoms have a greater potential to bioaccumulate, and are less likely to be degraded through metabolism than less chlorinated PCB congeners (Grant and Ross 2002). These highly chlorinated PCBs have also longer half – lives (Borgå et al. 2004).

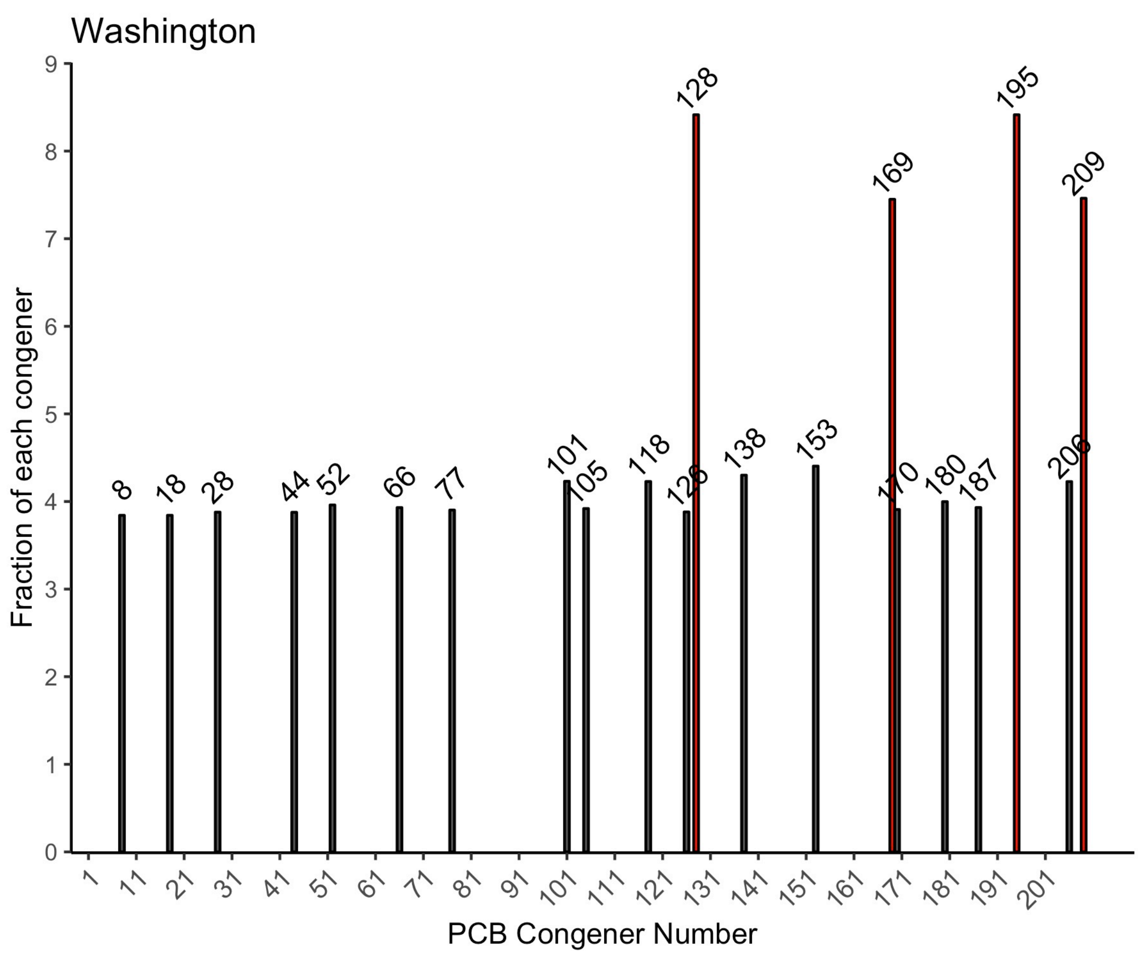

Figure 4.10. PCB composition patterns as the mean relative contribution (%) of PCB congeners to the total concentrations of PCBs in sediment samples collected in the US SRKW Critical Habitat (Juan de Fuca Strait and Puget Sound) from 2010 to 2016.

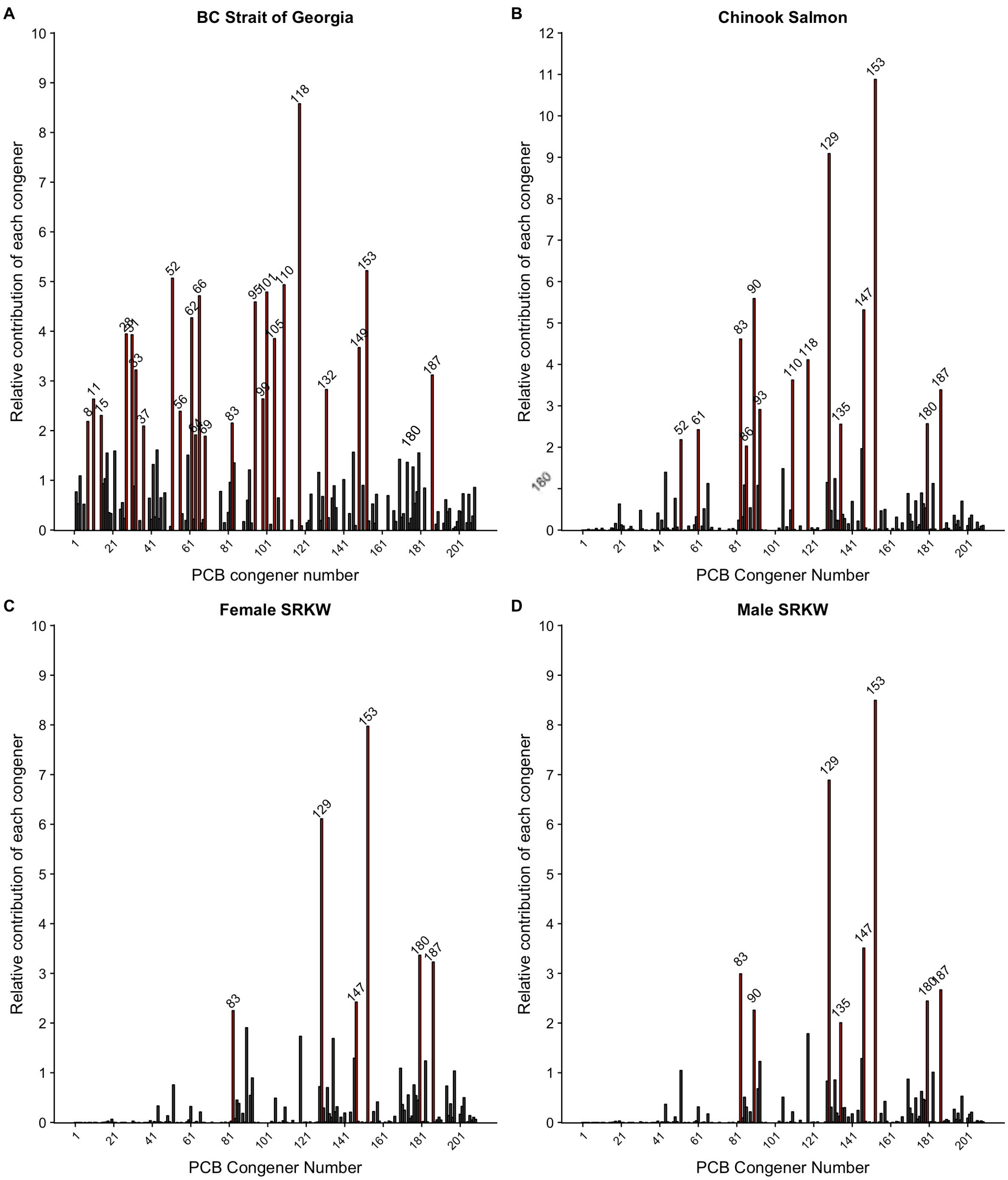

Figure 4.11. PCB composition patterns as the mean relative contribution (%) of PCB congeners to the total concentrations of PCBs in sediment samples collected in the Strait of Georgia (including BC SRKW Critical Habitat) from 2010 to 2017 (A), Chinook salmon collected in 2014 (B), female SRKW (C) and male SRKW (D) collected in 2015.

Chapter 5.

Conclusion

The contribution of PCBs stored in local sediment in the Salish Sea to the bioaccumulation of PCBs in Chinook salmon and SRKW was investigated in this research project. The results suggest that PCBs in the marine food web of SRKW originate to a significant extent from local environmental sources in the Salish Sea. Several lines of evidence provide support for this assertion including: i) the similarity of the temporal trends of the concentrations of PCBs in sediments, Chinook salmon and SRKW; ii) the presence of PCBs throughout the SRKW Critical Habitat at levels greater than the sediment concentrations recommended to be protective of SRKW; iii) the BSAF modelling indicates the concentrations of PCBs in sediment could bioaccumulate to the levels of PCBs observed in SRKW; and iv) the similarity in the PCB congener patterns in sediments, Chinook salmon and SRKW.

The concentrations of PCBs in sediment in the Salish Sea have not declined substantially in the 15 year period from 2002 to 2017. Through natural processes of sediment burial, the half – life of PCBs in the Salish Sea is estimated to be 10 years. Therefore, in 15 years, the concentration of PCBs in sediment would be expected to decrease by more than half. Since the decline in concentrations of PCBs in sediment in the Salish Sea was not observed, these results suggest an ongoing regional input of PCBs into the Salish Sea marine environment including the Critical Habitat of SRKW. Furthermore, the concentrations of PCBs in SRKW have not declined over two decades in female or male SRKW despite half – lives of 2 and 12 years, respectively. This suggests that SRKW continue to bioaccumulate PCBs through consumption of contaminated prey.

Given the half – lives of PCBs in Chinook salmon are on average 1.5 years and a portion of the population resides in the Salish Sea year-round, it is reasonable to expect that the PCB concentration in Chinook salmon reflects local PCB contamination. With biomagnification, the concentrations of PCBs taken up by biota from sediment can lead to concentration of PCBs in SRKW that are orders of magnitude higher. This relationship is formalized by the results of the food web bioaccumulation modelling using the BSAF that indicated that the local concentrations of PCBs in sediment in the Salish Sea were sufficient to produce the concentrations of PCBs that have been observed in adult male and female SRKW. Although the PCB composition pattern in local sediment is not the same as the PCB composition pattern observed in Chinook salmon and SRKW, there are several PCB congeners that exhibit a similar pattern. This provides further evidence to suggest that a fraction of the PCBs in SRKW could originate from local environmental sources.

The implication of the study results is that it is worthwhile to dedicate resources to local initiatives and proactive pollutant management actions that reduce PCB contamination in the Salish Sea and identify ongoing inputs of PCBs into the Salish Sea. Although global sources of PCBs contribute to the PCB contaminations of the Salish Sea environment and the SRKW food web, these cannot be controlled with provincial, state and national policy tools. Given the current decline of the SRKW population, efforts should be focussed in actions that can be taken within a short time frame to reduce the exposure of SRKW and their food web to PCBs in the Salish Sea. Research efforts should be targeted at filling knowledge gaps that could elucidate sources of PCBs such as additional measuring of concentrations of PCBs in Chinook salmon and using fingerprinting techniques to trace contaminants to specific habitat areas.

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Appendix A. Tables

Question 1: What are the concentrations of PCBs in sediments of SRKW habitats, Chinook salmon and SRKW? How do they differ spatially and temporally?

Table A 1 **Tukey HSD results from simultaneous multiple comparisons of the log 10 transformed mean concentrations of ∑PCB (pg/g dw) in sediment in the Strait of Georgia and SRKW Critical Habitat in BC.**

Year 1		Year 2	Estimate	Std. Error	t value	Pr(>|t|)	Significance
2003	-	2002	0.152698	0.362096	0.422	1	
2007	-	2002	0.151095	0.299844	0.504	1	
2010	-	2002	0.433933	0.278294	1.559	0.8314	
2011	-	2002	0.252367	0.308796	0.817	0.9975	
2013	-	2002	0.50766	0.276461	1.836	0.6564	
2014	-	2002	-0.757481	0.285539	-2.653	0.1572	
2015	-	2002	0.268018	0.277064	0.967	0.9913	
2016	-	2002	0.061179	0.298278	0.205	1	
2017	-	2002	0.28841	0.277236	1.04	0.9854	
2007	-	2003	-0.001603	0.266788	-0.006	1	
2010	-	2003	0.281236	0.242316	1.161	0.9695	
2011	-	2003	0.09967	0.276812	0.36	1	
2013	-	2003	0.354962	0.240208	1.478	0.8722	
2014	-	2003	-0.910179	0.com250604	-3.632	<0.01	**
2015	-	2003	0.11532	0.240902	0.479	1	
2016	-	2003	-0.091519	0.265027	-0.345	1	
2017	-	2003	0.135713	0.2411	0.563	0.9999	
2010	-	2007	0.282838	0.132325	2.137	0.4386	
2011	-	2007	0.101272	0.188196	0.538	0.9999	
2013	-	2007	0.356565	0.128425	2.776	0.1172	
2014	-	2007	-0.908576	0.146953	-6.183	<0.01	***
2015	-	2007	0.116923	0.129718	0.901	0.9948	
2016	-	2007	-0.089916	0.170389	-0.528	0.9999	
2017	-	2007	0.137315	0.130085	1.056	0.9838	
2011	-	2010	-0.181566	0.151521	-1.198	0.9624	
2013	-	2010	0.073727	0.063513	1.161	0.9695	
2014	-	2010	-1.191414	0.095584	-12.465	<0.01	***
2015	-	2010	-0.165915	0.066089	-2.51	0.2172	

Year 1		Year 2	Estimate	Std. Error	t value	Pr(>|t|)	Significance
2016	-	2010	-0.372754	0.128737	-2.895	0.0851	
2017	-	2010	-0.145523	0.066806	-2.178	0.4111	
2013	-	2011	0.255293	0.148128	1.723	0.7334	
2014	-	2011	-1.009848	0.164451	-6.141	<0.01	***
2015	-	2011	0.015651	0.14925	0.105	1	
2016	-	2011	-0.191188	0.185691	-1.03	0.9864	
2017	-	2011	0.036043	0.149569	0.241	1	
2014	-	2013	-1.265141	0.090107	-14.04	<0.01	***
2015	-	2013	-0.239642	0.057886	-4.14	<0.01	**
2016	-	2013	-0.446481	0.124725	-3.58	0.0102	*
2017	-	2013	-0.21925	0.058704	-3.735	<0.01	**
2015	-	2014	1.025499	0.091941	11.154	<0.01	***
2016	-	2014	0.81866	0.143731	5.696	<0.01	***
2017	-	2014	1.045891	0.092458	11.312	<0.01	***
2016	-	2015	-0.206839	0.126056	-1.641	0.7853	
2017	-	2015	0.020392	0.061482	0.332	1	
2017	-	2016	0.227231	0.126434	1.797	0.6841	

Significance codes: 0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1 (Adjusted p values reported -- single-step method)

Table A 2 Tukey HSD results from simultaneous multiple comparisons of the log 10 transformed mean concentrations of ∑PCB (pg/g dw) in sediment in US SRKW Critical Habitat in Juan de Fuca Strait and Puget Sound.

Year 1		Year 2	Estimate	Std. Error	t value	Pr(>|t|)	Significance
2003	-	2002	0.049412	0.031095	1.589	0.9478	
2004	-	2002	0.3466	0.033367	10.387	<0.01	***
2005	-	2002	0.485332	0.033367	14.545	<0.01	***
2006	-	2002	0.124715	0.031095	4.011	<0.01	**
2007	-	2002	0.01662	0.031095	0.535	1	
2008	-	2002	0.066319	0.033367	1.988	0.7722	
2009	-	2002	0.181744	0.02985	6.088	<0.01	***
2010	-	2002	0.189164	0.033971	5.568	<0.01	***
2011	-	2002	0.357155	0.030918	11.552	<0.01	***
2012	-	2002	0.368586	0.031095	11.854	<0.01	***
2013	-	2002	0.362887	0.031095	11.67	<0.01	***
2014	-	2002	0.353397	0.030584	11.555	<0.01	***
2016	-	2002	0.392878	0.028256	13.904	<0.01	***
2004	-	2003	0.297189	0.033367	8.907	<0.01	***
2005	-	2003	0.435921	0.033367	13.064	<0.01	***
2006	-	2003	0.075303	0.031095	2.422	0.4617	
2007	-	2003	-0.032791	0.031095	-1.055	0.9988	
2008	-	2003	0.016907	0.033367	0.507	1	
2009	-	2003	0.132332	0.02985	4.433	<0.01	***
2010	-	2003	0.139753	0.033971	4.114	<0.01	**
2011	-	2003	0.307743	0.030918	9.954	<0.01	***
2012	-	2003	0.319174	0.031095	10.265	<0.01	***
2013	-	2003	0.313475	0.031095	10.081	<0.01	***
2014	-	2003	0.303985	0.030584	9.94	<0.01	***
2016	-	2003	0.343466	0.028256	12.155	<0.01	***
2005	-	2004	0.138732	0.035495	3.909	<0.01	**
2006	-	2004	-0.221886	0.033367	-6.65	<0.01	***
2007	-	2004	-0.32998	0.033367	-9.889	<0.01	***
2008	-	2004	-0.280281	0.035495	-7.896	<0.01	***
2009	-	2004	-0.164857	0.032211	-5.118	<0.01	***
2010	-	2004	-0.157436	0.036063	-4.366	<0.01	**
2011	-	2004	0.010554	0.033202	0.318	1	
2012	-	2004	0.021985	0.033367	0.659	1	
2013	-	2004	0.016286	0.033367	0.488	1	
2014	-	2004	0.006796	0.032891	0.207	1	
2016	-	2004	0.046277	0.030739	1.505	0.966	
2006	-	2005	-0.360618	0.033367	-10.808	<0.01	***
2007	-	2005	-0.468712	0.033367	-14.047	<0.01	***
2008	-	2005	-0.419013	0.035495	-11.805	<0.01	***
2009	-	2005	-0.303589	0.032211	-9.425	<0.01	***
2010	-	2005	-0.296168	0.036063	-8.213	<0.01	***
2011	-	2005	-0.128178	0.033202	-3.861	<0.01	**
2012	-	2005	-0.116746	0.033367	-3.499	0.0334	*
2013	-	2005	-0.122445	0.033367	-3.67	0.0184	*

Year 1		Year 2	Estimate	Std. Error	t value	Pr(>|t|)	Significance
2014	-	2005	-0.131935	0.032891	-4.011	<0.01	**
2016	-	2005	-0.092454	0.030739	-3.008	0.1369	
2007	-	2006	-0.108094	0.031095	-3.476	0.035	*
2008	-	2006	-0.058396	0.033367	-1.75	0.8954	
2009	-	2006	0.057029	0.02985	1.91	0.8179	
2010	-	2006	0.06445	0.033971	1.897	0.8256	
2011	-	2006	0.23244	0.030918	7.518	<0.01	***
2012	-	2006	0.243871	0.031095	7.843	<0.01	***
2013	-	2006	0.238172	0.031095	7.66	<0.01	***
2014	-	2006	0.228682	0.030584	7.477	<0.01	***
2016	-	2006	0.268163	0.028256	9.49	<0.01	***
2008	-	2007	0.049699	0.033367	1.489	0.9688	
2009	-	2007	0.165123	0.02985	5.532	<0.01	***
2010	-	2007	0.172544	0.033971	5.079	<0.01	***
2011	-	2007	0.340534	0.030918	11.014	<0.01	***
2012	-	2007	0.351966	0.031095	11.319	<0.01	***
2013	-	2007	0.346266	0.031095	11.136	<0.01	***
2014	-	2007	0.336777	0.030584	11.012	<0.01	***
2016	-	2007	0.376258	0.028256	13.316	<0.01	***
2009	-	2008	0.115425	0.032211	3.583	0.0251	*
2010	-	2008	0.122845	0.036063	3.406	0.0429	*
2011	-	2008	0.290836	0.033202	8.76	<0.01	***
2012	-	2008	0.302267	0.033367	9.059	<0.01	***
2013	-	2008	0.296568	0.033367	8.888	<0.01	***
2014	-	2008	0.287078	0.032891	8.728	<0.01	***
2016	-	2008	0.326559	0.030739	10.623	<0.01	***
2010	-	2009	0.007421	0.032836	0.226	1	
2011	-	2009	0.175411	0.029666	5.913	<0.01	***
2012	-	2009	0.186842	0.02985	6.259	<0.01	***
2013	-	2009	0.181143	0.02985	6.068	<0.01	***
2014	-	2009	0.171653	0.029318	5.855	<0.01	***
2016	-	2009	0.211134	0.026881	7.854	<0.01	***
2011	-	2010	0.16799	0.033809	4.969	<0.01	***
2012	-	2010	0.179422	0.033971	5.282	<0.01	***
2013	-	2010	0.173723	0.033971	5.114	<0.01	***
2014	-	2010	0.164233	0.033504	4.902	<0.01	***
2016	-	2010	0.203714	0.031393	6.489	<0.01	***
2012	-	2011	0.011431	0.030918	0.37	1	
2013	-	2011	0.005732	0.030918	0.185	1	
2014	-	2011	-0.003758	0.030403	-0.124	1	
2016	-	2011	0.035723	0.028061	1.273	0.9921	
2013	-	2012	-0.005699	0.031095	-0.183	1	
2014	-	2012	-0.015189	0.030584	-0.497	1	
2016	-	2012	0.024292	0.028256	0.86	0.9999	
2014	-	2013	-0.00949	0.030584	-0.31	1	
2016	-	2013	0.029991	0.028256	1.061	0.9987	
2016	-	2014	0.039481	0.027693	1.426	0.9783	

Significance codes: 0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1 (Adjusted p values reported -- single-step method)

Table A 3 Tukey HSD results from simultaneous multiple comparisons of the log 10 transformed mean concentrations of ∑PCB (mg/kg lw) in female SRKW.

Year 1		Year 2	Estimate	Std. Error	t value	Pr(>|t|)
2006	-	1996	-0.05361	0.6019	-0.089	1
2007	-	1996	-0.4574	0.40127	-1.14	0.663
2015	-	1996	-0.69548	0.40127	-1.733	0.341
2007	-	2006	-0.40379	0.53082	-0.761	0.864
2015	-	2006	-0.64187	0.53082	-1.209	0.622
2015	-	2007	-0.23808	0.28374	-0.839	0.828

Table A 4 Tukey HSD results from simultaneous multiple comparisons of the log 10 transformed mean concentrations of ∑PCB (mg/kg lw) in male SRKW.

Year 1		Year 2	Estimate	Std. Error	t value	Pr(>|t|)
1996	-	1993	0.550724	0.374047	1.472	0.744
2000	-	1993	0.903167	0.458112	1.971	0.462
2004	-	1993	-0.144675	0.305408	-0.474	0.999
2006	-	1993	0.401631	0.323934	1.24	0.858
2007	-	1993	-0.002903	0.341457	-0.009	1
2015	-	1993	-0.478448	0.458112	-1.044	0.928
2000	-	1996	0.352443	0.458112	0.769	0.983
2004	-	1996	-0.695399	0.305408	-2.277	0.315
2006	-	1996	-0.149094	0.323934	-0.46	0.999
2007	-	1996	-0.553628	0.341457	-1.621	0.66
2015	-	1996	-1.029172	0.458112	-2.247	0.328
2004	-	2000	-1.047842	0.404017	-2.594	0.201
2006	-	2000	-0.501536	0.418197	-1.199	0.875
2007	-	2000	-0.90607	0.431912	-2.098	0.397
2015	-	2000	-1.381614	0.528982	-2.612	0.195
2006	-	2004	0.546306	0.241446	2.263	0.321
2007	-	2004	0.141772	0.264491	0.536	0.997
2015	-	2004	-0.333773	0.404017	-0.826	0.975
2007	-	2006	-0.404534	0.285683	-1.416	0.774
2015	-	2006	-0.880078	0.418197	-2.104	0.394
2015	-	2007	-0.475544	0.431912	-1.101	0.911

Question 2: Can the concentrations of PCBs in the habitat of SRKWs contribute significantly to the concentration of PCBs observed in Chinook salmon and SRKW?

Table A 5 Biota – sediment accumulation factors (BSAF) for the Salish Sea and estimated concentrations of ∑PCB in Chinook salmon, male SRKW and female SRKW.

Species	Gender	Time Spent (%)	Log BSAF	SD Log BSAF	Concentration of PCBs (mg/kg lw)	Upper SD concentration of PCBs (mg/kg lw)	Lower SD concentration of PCBs (mg/kg lw)
Chinook	NA	0.29	1.36	0.18	1.24	1.88	0.82
SRKW	Male	0.63	4.10	0.33	195.93	418.90	91.64
SRKW	Female	0.63	3.35	0.33	34.84	74.49	16.30

The log BSAF values were adjusted for time spent by Chinook salmon and SRKW in the Salish Sea region (see Equation 2).

Table A 6 Estimated half – lives (years) of PCB congeners in Chinook salmon, adult male SRKW, adult female SRKW and juvenile SRKW.

PCB Congener CAS #	log Kow (unitless)	Half - life (years) Chinook salmon		Half - life (years) - SRKW		
		BC SRKW Critical Habitat and Strait of Georgia	US SRKW Critical Habitat	Adult Male	Adult Female	Juvenile
8	5.27	0.46	0.61	8.96	1.52	2.58
18	5.46	0.57	0.69	8.88	1.51	2.57
28	5.89	0.74	0.81	10.57	1.57	2.76
31	5.82	0.79	0.84	10.77	1.58	2.78
33	5.99	0.74	0.81	10.79	1.58	2.78
44	5.82	0.81	0.85	11.41	1.6	2.84
49	6.09	0.85	0.88	11.12	1.59	2.81
52	6.08	0.84	0.87	11.15	1.59	2.82
56	6.35	0.88	0.89	11.51	1.6	2.85
60	6.35	0.9	0.91	11.61	1.6	2.86
66	6.44	0.87	0.89	11.61	1.6	2.86
70	6.44	0.9	0.91	11.54	1.6	2.85
74	6.44	0.9	0.91	11.58	1.6	2.86
87	6.39	0.96	0.95	11.6	1.6	2.86
95	6.39	0.89	0.9	11.55	1.6	2.85
99	6.65	0.96	0.96	11.61	1.6	2.86
101	6.64	0.95	0.95	11.54	1.6	2.86
105	6.91	1.11	1.09	11.65	1.6	2.86
110	6.74	0.95	0.95	11.6	1.6	2.86
118	6.91	1.03	1.01	11.63	1.6	2.86
128	7.02	1.1	1.08	11.67	1.6	2.86
132	6.86	1.01	0.99	11.65	1.6	2.86
138	7.11	1.3	1.27	11.68	1.6	2.86
141	7.1	1.08	1.06	11.67	1.6	2.86
149	6.95	1.03	1.01	11.65	1.6	2.86
151	6.92	1.02	1.01	11.65	1.6	2.86
153	7.18	1.14	1.11	11.64	1.6	2.86
156	7.46	1.18	1.15	11.69	1.6	2.87
158	7.46	1.12	1.09	11.68	1.6	2.87
170	7.57	1.28	1.25	11.69	1.6	2.87
174	7.41	1.21	1.18	11.68	1.6	2.87
177	7.38	1.19	1.17	11.68	1.6	2.87
180	7.66	1.28	1.25	11.69	1.6	2.87
183	7.5	1.25	1.22	11.9	1.6	2.87

PCB Congener CAS #	log Kow (unitless)	Half - life (years) Chinook salmon		Half - life (years) - SRKW		
		BC SRKW Critical Habitat and Strait of Georgia	US SRKW Critical Habitat	Adult Male	Adult Female	Juvenile
187	7.47	1.24	1.21	11.69	1.6	2.87
194	8.12	1.68	1.66	11.75	1.6	2.87
195	7.88	1.47	1.44	11.71	1.6	2.87
201	7.94	1.5	1.48	11.72	1.6	2.87
203	7.95	1.52	1.5	11.72	1.6	2.87

Question 3: PCB congener patterns in sediment, Chinook salmon and SRKW

Table A 7 Relative contribution of each PCB congener to ∑PCB in Strait of Georgia and BC SRKW Critical Habitat.

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
1	0.70	0.48	107	0.65	0.23
2	0.77	0.83	110	4.94	1.62
3	0.53	0.50	114	0.20	0.14
4	1.09	0.78	118	8.58	3.04
6	0.52	0.48	119	0.08	0.03
8	2.18	1.51	122	0.15	0.15
11	2.63	2.02	123	0.19	0.15
15	2.30	1.15	124	0.72	0.22
16	0.93	0.58	128	1.16	0.36
17	1.03	0.58	129	0.19	0.09
18	1.55	0.81	130	0.67	0.31
19	0.35	0.33	132	2.83	1.37
20	0.33	0.19	133	0.25	0.29
22	1.59	0.82	135	0.64	0.14
25	0.42	0.21	136	0.89	0.51
26	0.55	0.20	137	0.45	0.44
27	0.24	0.19	141	1.01	0.61
28	3.95	1.33	144	0.33	0.24
31	3.93	1.80	146	1.57	0.64
32	0.88	0.37	147	0.09	0.04
33	3.22	1.22	149	3.67	0.70
35	0.19	0.15	151	0.90	0.20
37	2.09	0.77	153	5.22	1.08
40	0.64	0.21	154	0.18	0.20
41	0.21	0.28	156	0.52	0.15
42	1.32	0.47	157	0.14	0.05
43	0.26	0.35	158	0.72	0.37
44	1.61	0.33	164	0.69	0.27
45	0.23	0.12	167	0.39	0.23
46	0.65	0.76	168	0.17	0.16
48	0.75	0.34	170	1.42	0.87

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
51	0.07	0.05	171	0.26	0.07
52	5.07	1.94	172	0.33	0.30
56	2.39	0.79	174	1.36	0.87
57	0.33	0.51	175	0.13	0.19
59	0.19	0.06	176	0.24	0.15
60	1.51	0.51	177	1.27	0.55
62	4.27	0.88	178	0.53	0.26
63	0.22	0.46	179	0.77	0.40
64	1.91	0.62	180	1.55	0.32
66	4.71	2.84	183	0.84	0.54
67	0.14	0.13	187	3.12	1.51
68	0.21	0.28	189	0.12	0.21
69	1.89	0.52	190	0.37	0.22
77	0.78	0.49	193	0.14	0.05
79	0.15	0.14	194	0.61	0.41
81	0.35	0.47	195	0.37	0.28
82	0.96	0.32	196	0.43	0.33
83	2.15	0.56	198	0.03	0.02
84	1.35	0.91	199	0.06	0.03
89	0.17	0.20	200	0.17	0.20
91	0.60	0.20	201	0.39	0.38
92	1.21	0.48	202	0.37	0.22
93	0.14	0.21	203	0.73	0.37
95	4.59	2.00	205	0.15	0.20
99	2.64	0.56	206	0.72	0.42
101	4.79	1.22	207	0.14	0.17
103	0.11	0.15	208	0.28	0.20
105	3.85	1.39	209	0.86	0.59

Table A 8 Relative contribution of each PCB congener to ∑PCB in US SRKW Critical Habitat.

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
8	3.84	0.10	128	8.41	0.02
18	3.84	0.10	138	4.30	0.17
28	3.88	0.09	153	4.40	0.18
44	3.88	0.10	169	7.45	0.19
52	3.96	0.12	170	3.91	0.11
66	3.93	0.11	180	4.00	0.13
77	3.90	0.11	187	3.93	0.12
101	4.23	0.17	195	8.41	0.02
105	3.92	0.11	206	4.23	0.06
118	4.23	0.16	209	7.46	0.18
126	3.88	0.10			

Table A 9 Relative contribution of each PCB congener to ∑PCB in Chinook salmon samples collected in 2014.

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
1	0.00	0.00	109	0.48	0.11
2	0.00	0.00	110	3.62	0.86
3	0.00	0.00	111	0.02	0.00
4	0.02	0.01	114	0.10	0.02
5	0.00	NA	118	4.11	0.96
6	0.01	0.00	120	0.05	0.01
7	0.00	0.00	121	0.02	0.01
8	0.04	0.02	122	0.01	0.01
9	0.00	0.00	123	0.05	0.01
10	0.00	0.00	126	0.01	0.00
11	0.04	0.03	127	0.01	0.00
12	0.00	0.00	128	1.15	0.27
15	0.00	0.00	129	9.09	2.02
16	0.05	0.03	130	0.47	0.10
17	0.04	0.02	131	0.02	0.01
18	0.16	0.08	132	1.24	0.28
19	0.01	0.01	133	0.24	0.05

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
20	0.63	0.27	134	0.23	0.05
21	0.13	0.07	135	2.56	0.55
22	0.10	0.05	136	0.38	0.09
23	0.00	0.00	137	0.28	0.07
24	0.00	0.00	139	0.15	0.03
25	0.03	0.01	141	0.69	0.16
26	0.09	0.04	144	0.22	0.05
27	0.01	0.01	145	0.00	0.00
31	0.48	0.22	146	1.96	0.41
32	0.03	0.01	147	5.32	1.17
34	0.01	0.00	148	0.05	0.01
37	0.01	0.00	150	0.02	0.00
38	0.00	NA	152	0.00	0.00
39	0.00	0.00	153	10.88	2.63
40	0.41	0.14	155	0.03	0.01
42	0.24	0.09	156	0.46	0.12
43	0.04	0.01	158	0.50	0.11
44	1.40	0.42	159	0.03	0.01
45	0.05	0.02	162	0.04	0.01
46	0.01	0.00	164	0.31	0.07
48	0.04	0.01	165	0.02	0.01
49	0.77	0.23	167	0.17	0.05
50	0.07	0.03	170	0.88	0.25
52	2.18	0.70	171	0.39	0.09
54	0.00	0.00	172	0.21	0.06
56	0.10	0.05	174	0.70	0.16
57	0.01	0.00	175	0.08	0.02
58	0.01	0.00	176	0.13	0.03
59	0.12	0.04	177	0.90	0.18
60	0.32	0.10	178	0.64	0.15
61	2.42	0.73	179	0.53	0.13
63	0.10	0.03	180	2.57	0.78
64	0.51	0.15	181	0.01	0.00
66	1.12	0.33	182	0.03	0.01
67	0.02	0.01	183	1.12	0.30
68	0.06	0.03	184	0.02	0.01

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
72	0.04	0.01	187	3.39	0.76
77	0.02	0.01	188	0.01	0.00
79	0.04	0.01	189	0.04	0.01
80	0.00	NA	190	0.17	0.05
82	0.24	0.07	191	0.04	0.01
83	4.61	1.10	192	0.00	NA
84	0.32	0.09	194	0.36	0.17
85	1.09	0.27	195	0.14	0.06
86	2.03	0.48	196	0.23	0.09
88	0.54	0.14	197	0.07	0.02
89	0.02	0.01	198	0.70	0.21
90	5.59	1.28	201	0.11	0.04
92	1.08	0.24	202	0.29	0.09
93	2.91	0.82	203	0.36	0.15
94	0.01	0.00	204	0.00	0.00
96	0.00	0.00	205	0.02	0.01
103	0.05	0.01	206	0.19	0.11
104	0.00	0.00	207	0.04	0.02
105	1.48	0.35	208	0.09	0.04
107	0.08	0.02	209	0.11	0.07

Table A 10 Relative contribution of each PCB congener to ∑PCB in adult female SRKW samples collected in 2015.

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
1	0.0003	0.0002	114	0.0388	0.0087
2	0.0002	0.0001	118	1.7335	0.3815
3	0.0005	0.0003	120	0.0062	0.0015
4	0.0008	0.0003	121	0.0070	0.0016
6	0.0001	0.0001	123	0.0121	0.0038
8	0.0014	0.0010	126	0.0016	0.0011
9	0.0000	0.0000	127	0.0099	0.0032
11	0.0019	0.0012	128	0.7206	0.1847
12	0.0001	0.0000	129	6.1115	0.7556
15	0.0005	0.0004	130	0.2903	0.0608
16	0.0042	0.0017	131	0.0115	0.0023
17	0.0074	0.0024	132	0.7008	0.3038
18	0.0259	0.0085	133	0.1753	0.0346
19	0.0008	0.0004	134	0.1038	0.0300
20	0.0639	0.0315	135	1.6898	0.4631
21	0.0027	0.0019	136	0.2151	0.0753
22	0.0015	0.0011	137	0.3145	0.0827
25	0.0008	0.0003	139	0.1046	0.0228
26	0.0016	0.0012	141	0.1897	0.0847
27	0.0008	0.0004	144	0.2066	0.0742
31	0.0259	0.0145	145	0.0007	0.0003
32	0.0011	0.0008	146	1.2927	0.1827
34	0.0002	0.0002	147	2.4227	0.6814
37	0.0005	0.0002	148	0.0225	0.0044
38	0.0002	0.0001	150	0.0096	0.0026
39	0.0005	0.0002	152	0.0010	0.0002
40	0.0349	0.0139	153	7.9727	0.6711
42	0.0179	0.0088	155	0.0107	0.0020
43	0.0071	0.0025	156	0.2190	0.0299
44	0.3318	0.0795	158	0.4125	0.1408
45	0.0119	0.0040	159	0.0250	0.0095
46	0.0014	0.0005	162	0.0058	0.0035
48	0.0139	0.0051	164	0.0279	0.0120
49	0.1343	0.0538	165	0.0112	0.0020
50	0.0150	0.0049	167	0.1231	0.0198

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
52	0.7559	0.2670	170	1.0902	0.2555
54	0.0001	0.0001	171	0.3611	0.1090
56	0.0016	0.0012	172	0.2465	0.0614
59	0.0142	0.0052	174	0.5541	0.2297
60	0.0499	0.0219	175	0.0785	0.0239
61	0.3206	0.0834	176	0.1292	0.0348
63	0.0018	0.0014	177	0.7559	0.2356
64	0.0200	0.0127	178	0.5402	0.1026
66	0.2105	0.0665	179	0.4478	0.1173
67	0.0007	NA	180	3.3654	1.0510
68	0.0024	0.0017	181	0.0123	0.0035
72	0.0051	0.0026	182	0.0196	0.0038
77	0.0009	0.0005	183	1.2379	0.3169
79	0.0065	0.0032	184	0.0118	0.0028
81	0.0006	0.0003	187	3.2281	0.6924
82	0.0250	0.0098	188	0.0104	0.0022
83	2.2498	0.6523	189	0.0524	0.0190
84	0.0786	0.0240	190	0.1048	0.0392
85	0.4494	0.1213	191	0.0478	0.0164
86	0.3810	0.1505	194	0.7326	0.4308
88	0.1826	0.0696	195	0.1397	0.0734
89	0.0056	0.0028	196	0.3744	0.1908
90	1.9062	0.4923	197	0.1000	0.0457
92	0.5451	0.1601	198	1.0353	0.4628
93	0.8953	0.2990	201	0.1645	0.0713
94	0.0027	0.0008	202	0.3232	0.1497
96	0.0013	0.0003	203	0.4982	0.2863
103	0.0209	0.0060	204	0.0022	0.0013
104	0.0003	0.0001	205	0.0140	0.0091
105	0.4890	0.1207	206	0.1379	0.1032
107	0.0023	0.0019	207	0.0488	0.0317
109	0.0338	0.0180	208	0.1023	0.0724
110	0.3081	0.1278	209	0.0608	0.0467
111	0.0028	0.0014			

Table A 11 Relative contribution of each PCB congener to ∑PCB in adult male SRKW samples collected in 2015.

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
1	0.0002	0.0002	114	0.0419	0.0087
2	0.0001	0.0001	118	1.7854	0.3815
3	0.0002	0.0003	120	0.0049	0.0015
4	0.0005	0.0003	121	0.0079	0.0016
6	0.0001	0.0001	123	0.0112	0.0038
7	0.0000	NA	127	0.0096	0.0032
8	0.0006	0.0010	128	0.8317	0.1847
9	0.0000	0.0000	129	6.8873	0.7556
11	0.0013	0.0012	130	0.3061	0.0608
12	0.0001	0.0000	131	0.0129	0.0023
15	0.0003	0.0004	132	0.8548	0.3038
16	0.0030	0.0017	133	0.1891	0.0346
17	0.0062	0.0024	134	0.1243	0.0300
18	0.0223	0.0085	135	2.0066	0.4631
19	0.0005	0.0004	136	0.2897	0.0753
20	0.0304	0.0315	137	0.2975	0.0827
21	0.0013	0.0019	139	0.1092	0.0228
22	0.0006	0.0011	141	0.1724	0.0847
25	0.0008	0.0003	144	0.2420	0.0742
26	0.0007	0.0012	145	0.0008	0.0003
27	0.0005	0.0004	146	1.2845	0.1827
31	0.0133	0.0145	147	3.5092	0.6814
32	0.0005	0.0008	148	0.0213	0.0044
34	0.0001	0.0002	150	0.0113	0.0026
37	0.0003	0.0002	152	0.0010	0.0002
38	0.0002	0.0001	153	8.4942	0.6711
39	0.0005	0.0002	155	0.0089	0.0020
40	0.0248	0.0139	156	0.1813	0.0299
42	0.0109	0.0088	158	0.4230	0.1408
43	0.0088	0.0025	159	0.0230	0.0095
44	0.3648	0.0795	162	0.0032	0.0035
45	0.0132	0.0040	164	0.0196	0.0120
46	0.0014	0.0005	165	0.0117	0.0020
48	0.0132	0.0051	167	0.1131	0.0198
49	0.1106	0.0538	170	0.8705	0.2555

PCB Congener CAS #	Mean fraction (%)	SD fraction (%)	PCB Congener CAS #	Mean fraction (%)	SD fraction (%)
50	0.0157	0.0049	171	0.2892	0.1090
52	1.0453	0.2670	172	0.1750	0.0614
54	0.0001	0.0001	174	0.4923	0.2297
56	0.0008	0.0012	175	0.0663	0.0239
59	0.0116	0.0052	176	0.1223	0.0348
60	0.0327	0.0219	177	0.6236	0.2356
61	0.3124	0.0834	178	0.4656	0.1026
63	0.0007	0.0014	179	0.4460	0.1173
64	0.0097	0.0127	180	2.4430	1.0510
66	0.1716	0.0665	181	0.0087	0.0035
68	0.0009	0.0017	182	0.0150	0.0038
72	0.0036	0.0026	183	1.0099	0.3169
77	0.0004	0.0005	184	0.0090	0.0028
79	0.0035	0.0032	186	0.0002	NA
82	0.0193	0.0098	187	2.6681	0.6924
83	2.9911	0.6523	188	0.0076	0.0022
84	0.0905	0.0240	189	0.0303	0.0190
85	0.5072	0.1213	190	0.0584	0.0392
86	0.3095	0.1505	191	0.0353	0.0164
88	0.2130	0.0696	194	0.2647	0.4308
89	0.0070	0.0028	195	0.0611	0.0734
90	2.2586	0.4923	196	0.1858	0.1908
92	0.6782	0.1601	197	0.0501	0.0457
93	1.2271	0.2990	198	0.5273	0.4628
94	0.0030	0.0008	201	0.0858	0.0713
96	0.0014	0.0003	202	0.1641	0.1497
103	0.0238	0.0060	203	0.2061	0.2863
104	0.0002	0.0001	204	0.0008	0.0013
105	0.5083	0.1207	205	0.0049	0.0091
107	0.0009	0.0019	206	0.0363	0.1032
109	0.0169	0.0180	207	0.0141	0.0317
110	0.2131	0.1278	208	0.0298	0.0724
111	0.0020	0.0014	209	0.0128	0.0467

Appendix B. Figures

Question 1: What are the concentrations of PCBs in sediments of SRKW habitats, Chinook salmon and SRKW? How do they differ spatially and temporally?

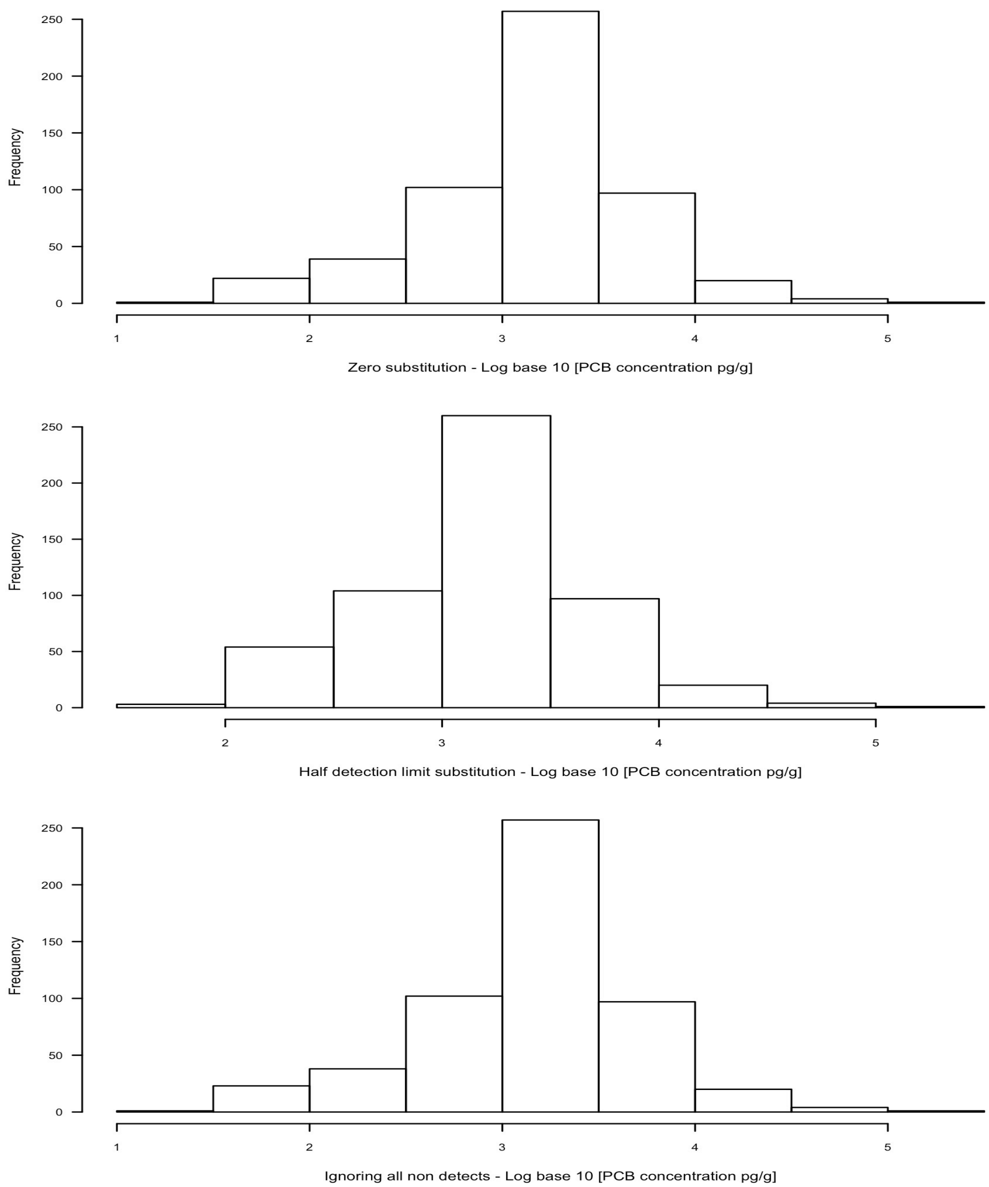


Figure B 1 **Histograms of log transformed concentrations of PCBs in sediment collected in the Strait of Georgia and in the BC SRKW Critical Habitat from 2010 to 2017. The histograms show the distribution of the data for each method of dealing with non – detects.**

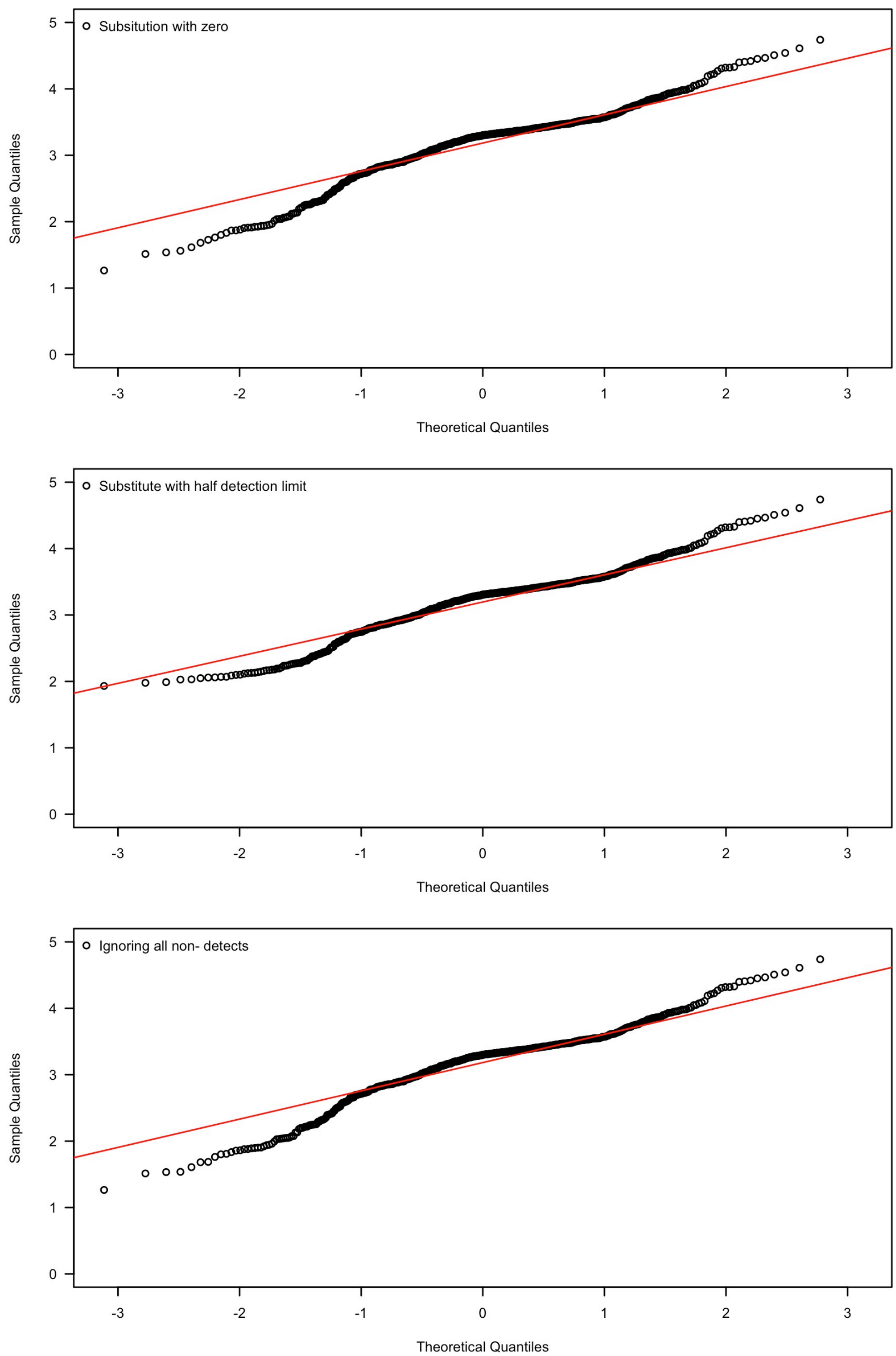


Figure B 2 **Normal quantile- quantile plots of log 10 transformed concentrations of ∑PCB in sediment collected in the Strait of Georgia and SRKW Critical Habitat in BC from 2010 to 2017. Each quantile – quantile plot shows a different method of dealing with non – detects. The straight line in red indicates a one to one relationship.**

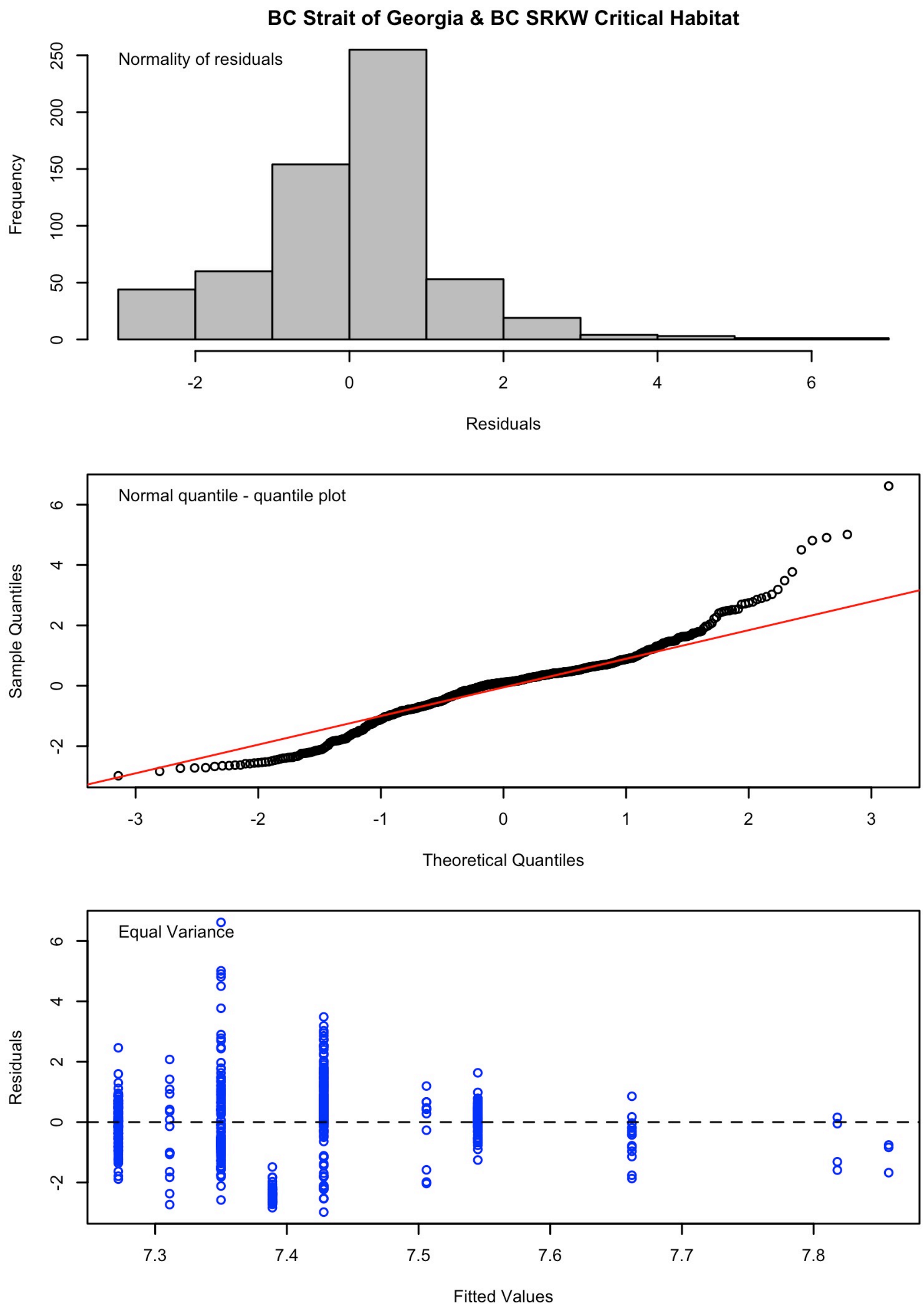


Figure B 3 **Diagnostic plots testing the assumptions of linear regression including a histogram of the one-way ANOVA residuals, normal quantile - quantile plot and equal variance plot of the residuals and fitted values for the log 10 transformed concentrations of ∑PCB in sediment collected in the BC Strait of Georgia and in the BC SRKW Critical Habitat from 1996 to 2017.**

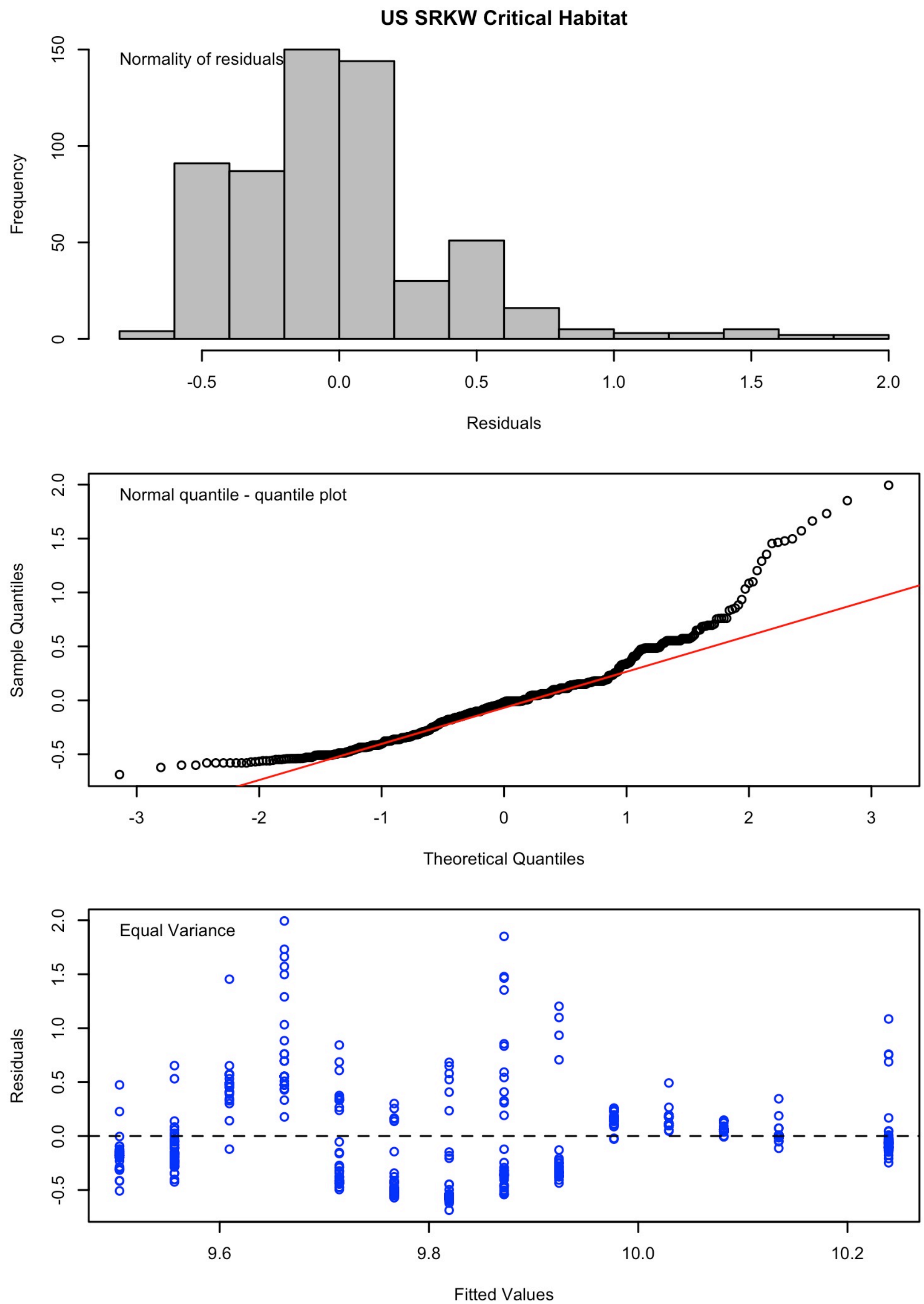


Figure B 4 **Diagnostic plots testing the assumptions of linear regression including a histogram of the one-way ANOVA residuals, normal quantile - quantile plot and equal variance plot of the residuals and fitted values for the log 10 transformed concentrations of ∑PCB in sediment collected in the US SRKW Critical Habitat from 2002 to 2016.**

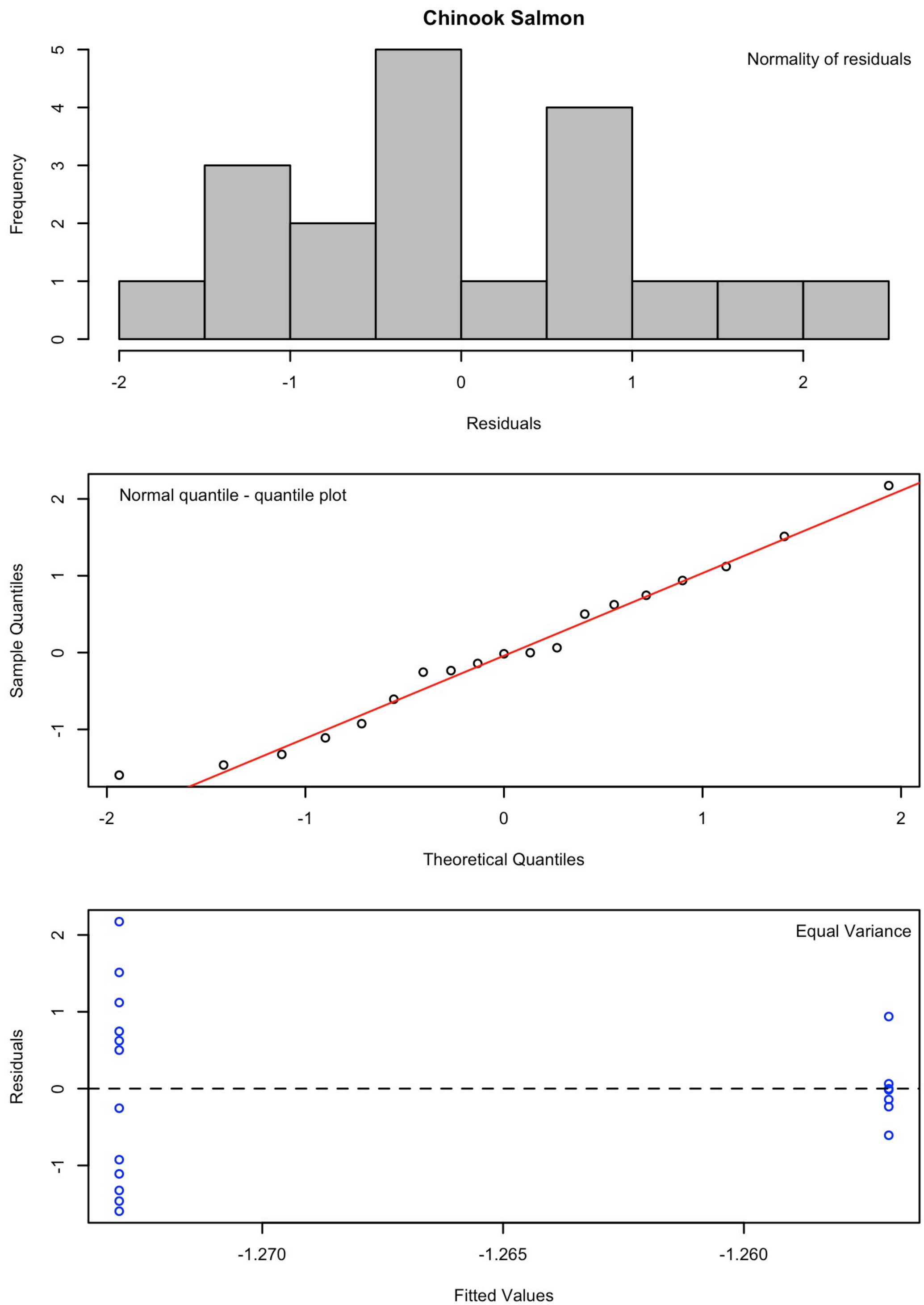


Figure B 5 Diagnostic plots testing the assumptions of linear regression including a histogram of the one-way ANOVA residuals, normal quantile - quantile plot and equal variance plot of the residuals and fitted values for the log 10 transformed concentrations of ∑PCB in Chinook salmon samples collected in 2010 and 2014.

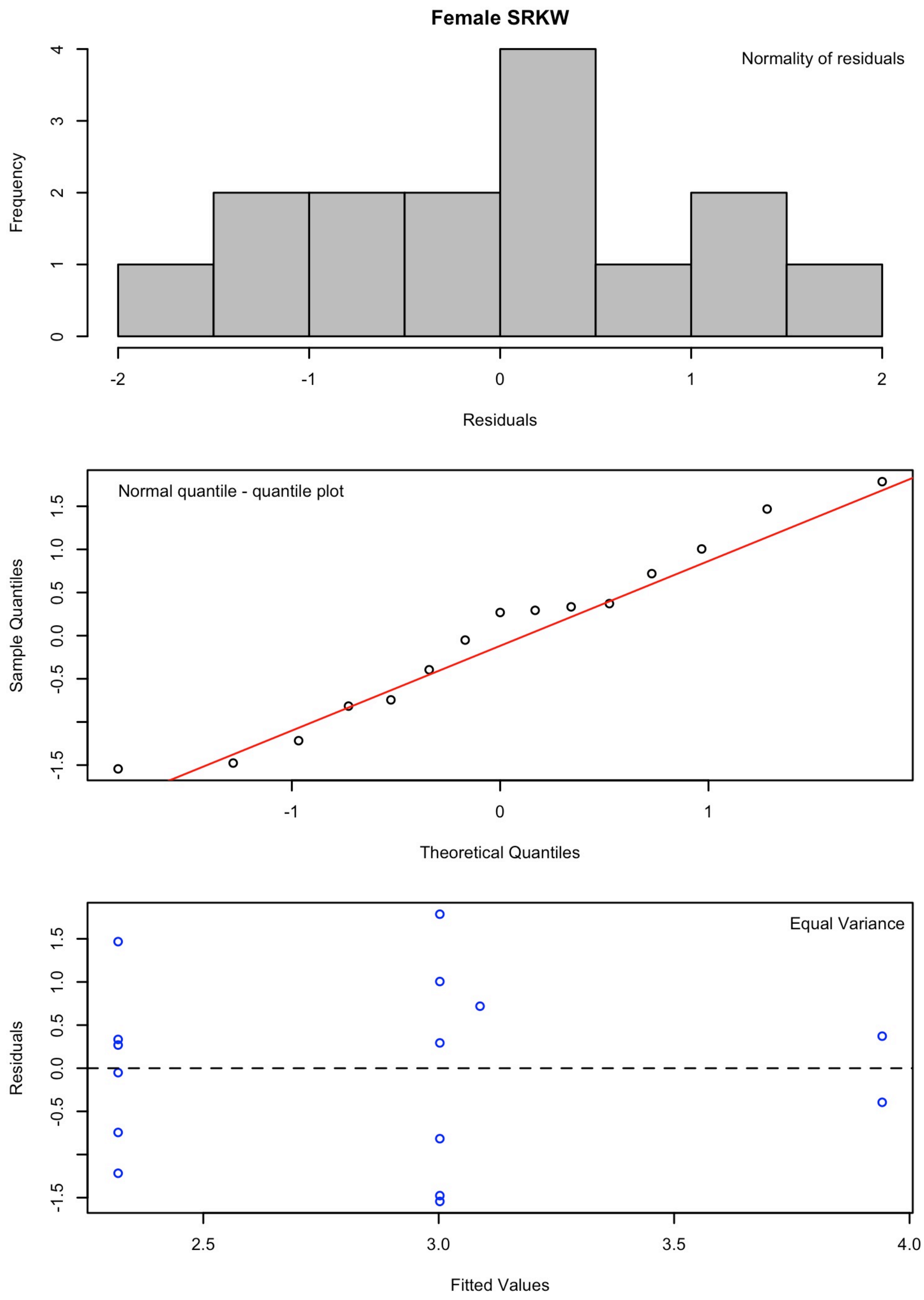


Figure B 6 **Diagnostic plots testing the assumptions of linear regression including a histogram of the one-way ANOVA residuals, normal quantile - quantile plot and equal variance plot of the residuals and fitted values for the log 10 transformed concentrations of ∑PCB in female SRKW samples collected from 1996 to 2015.**

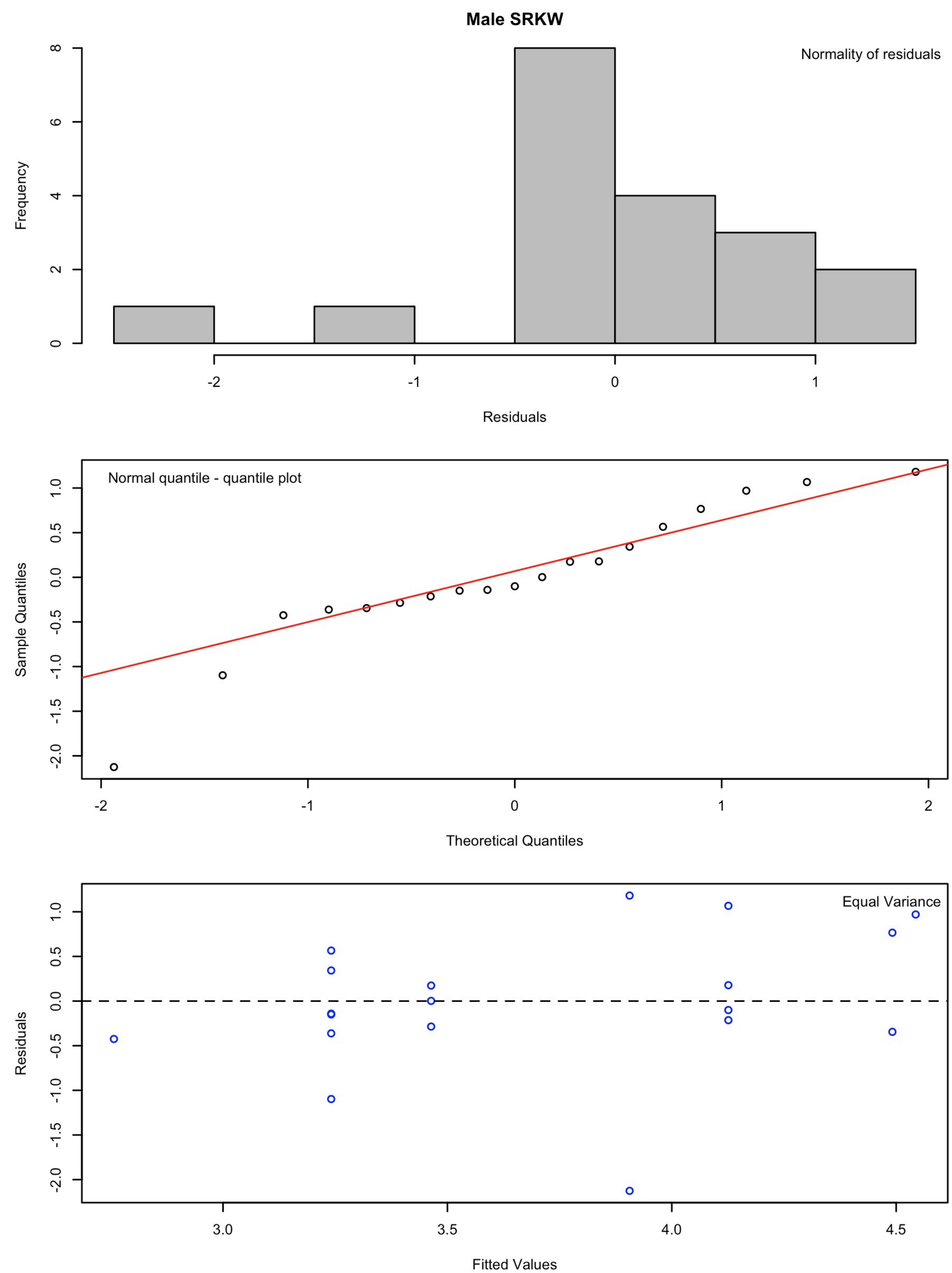
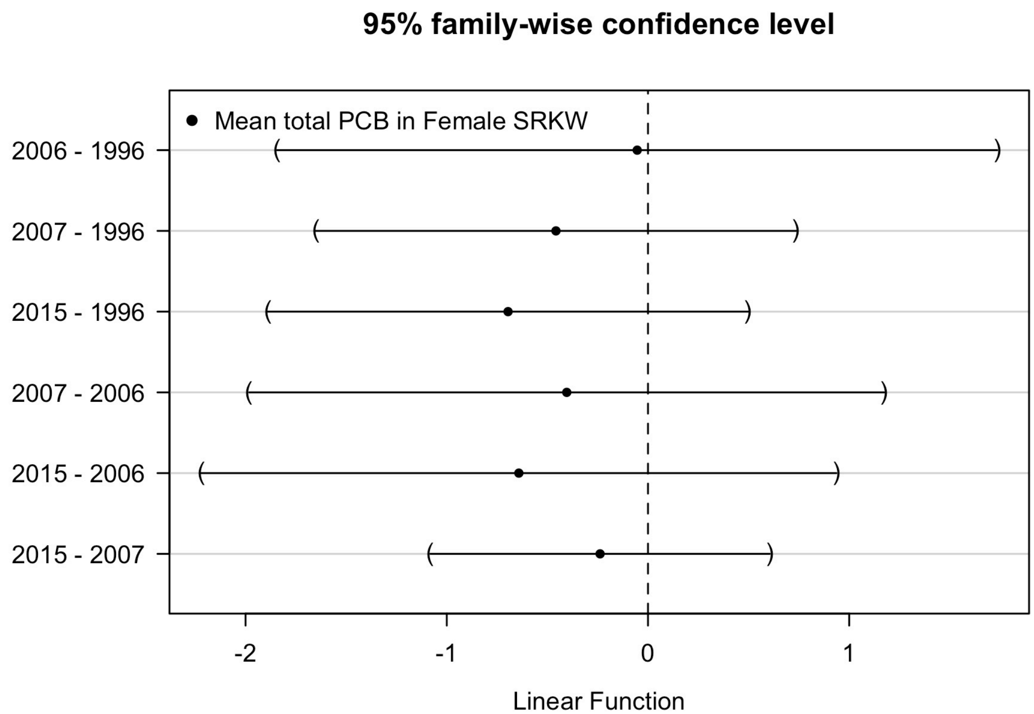


Figure B 7 **Diagnostic plots testing the assumptions of linear regression including a histogram of the one-way ANOVA residuals, normal quantile - quantile plot and equal variance plot of the residuals and fitted values for the log 10 transformed concentrations of ∑PCB in male SRKW samples collected from 1993 to 2015.**



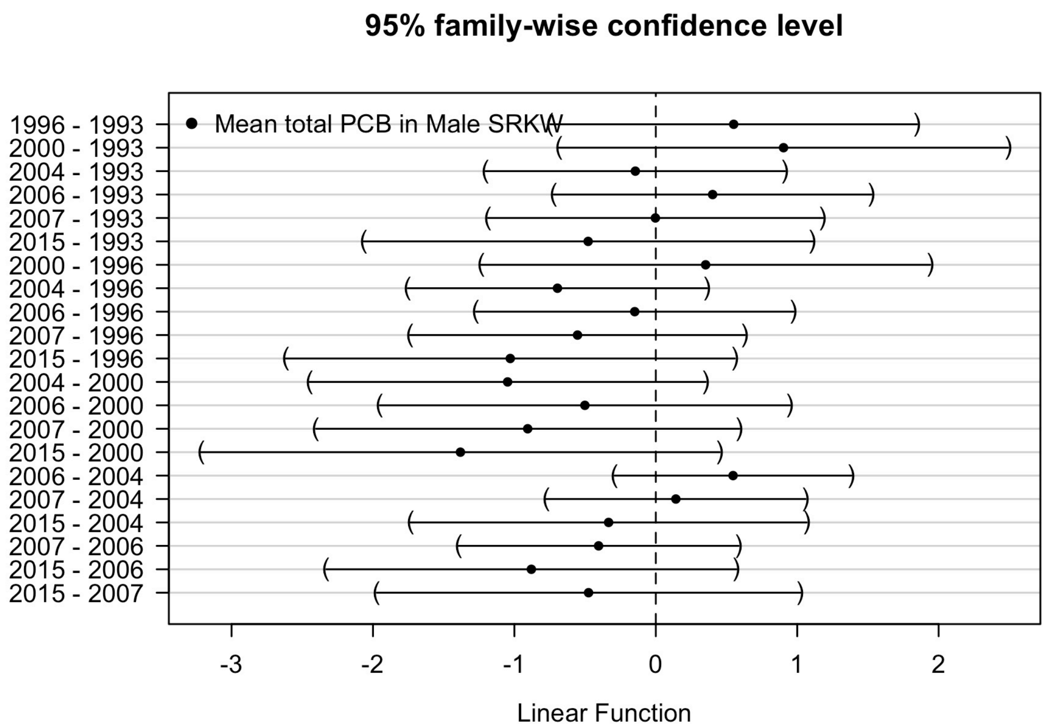


Figure B 8 Tukey Honest Significant Difference test results for comparisons of annual geometric mean ∑PCB concentration in female and male SRKW.

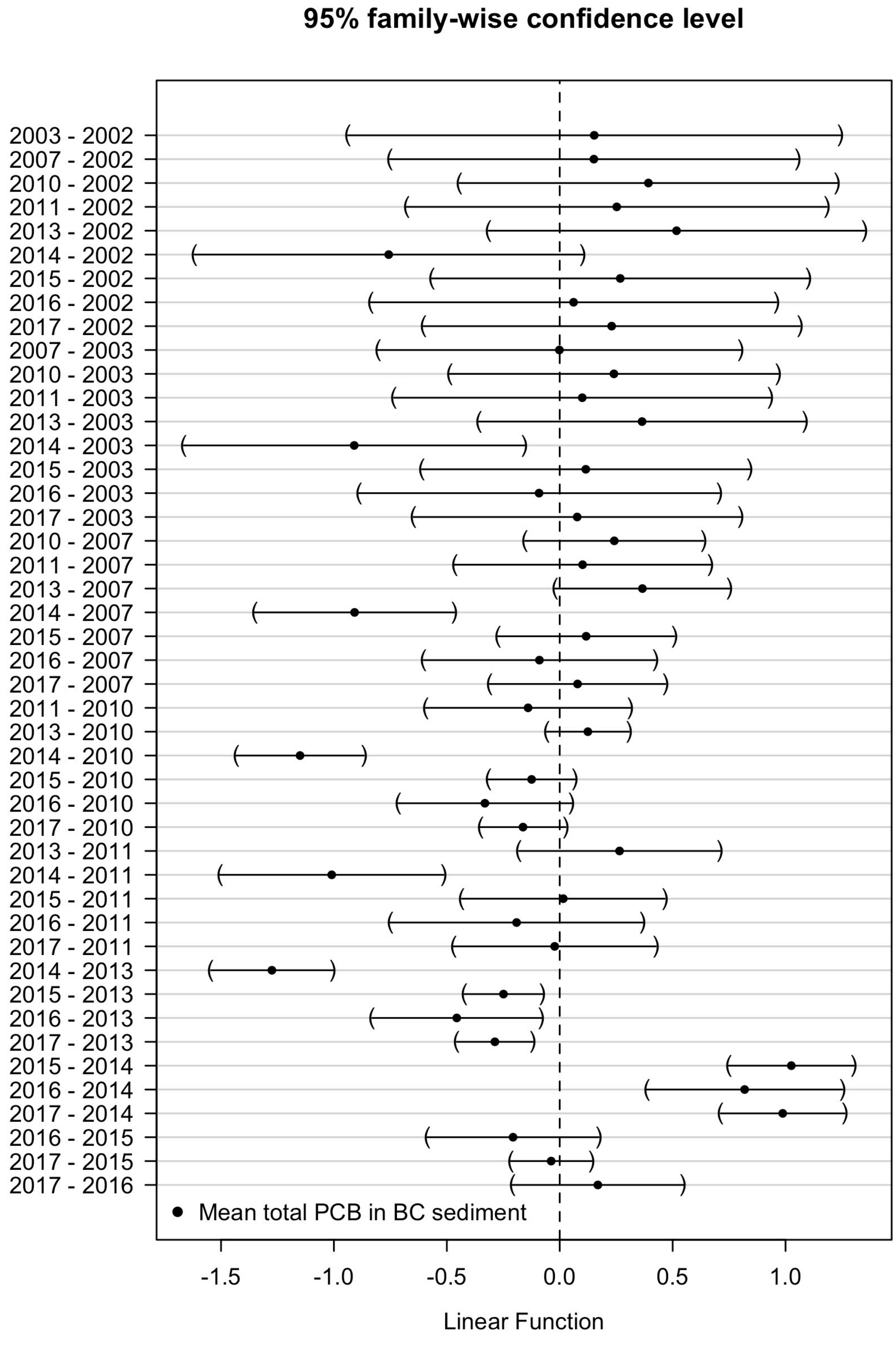


Figure B 9 **Tukey Honest Significant Difference test results for comparisons of annual geometric mean ∑PCB concentration in BC sediments.**

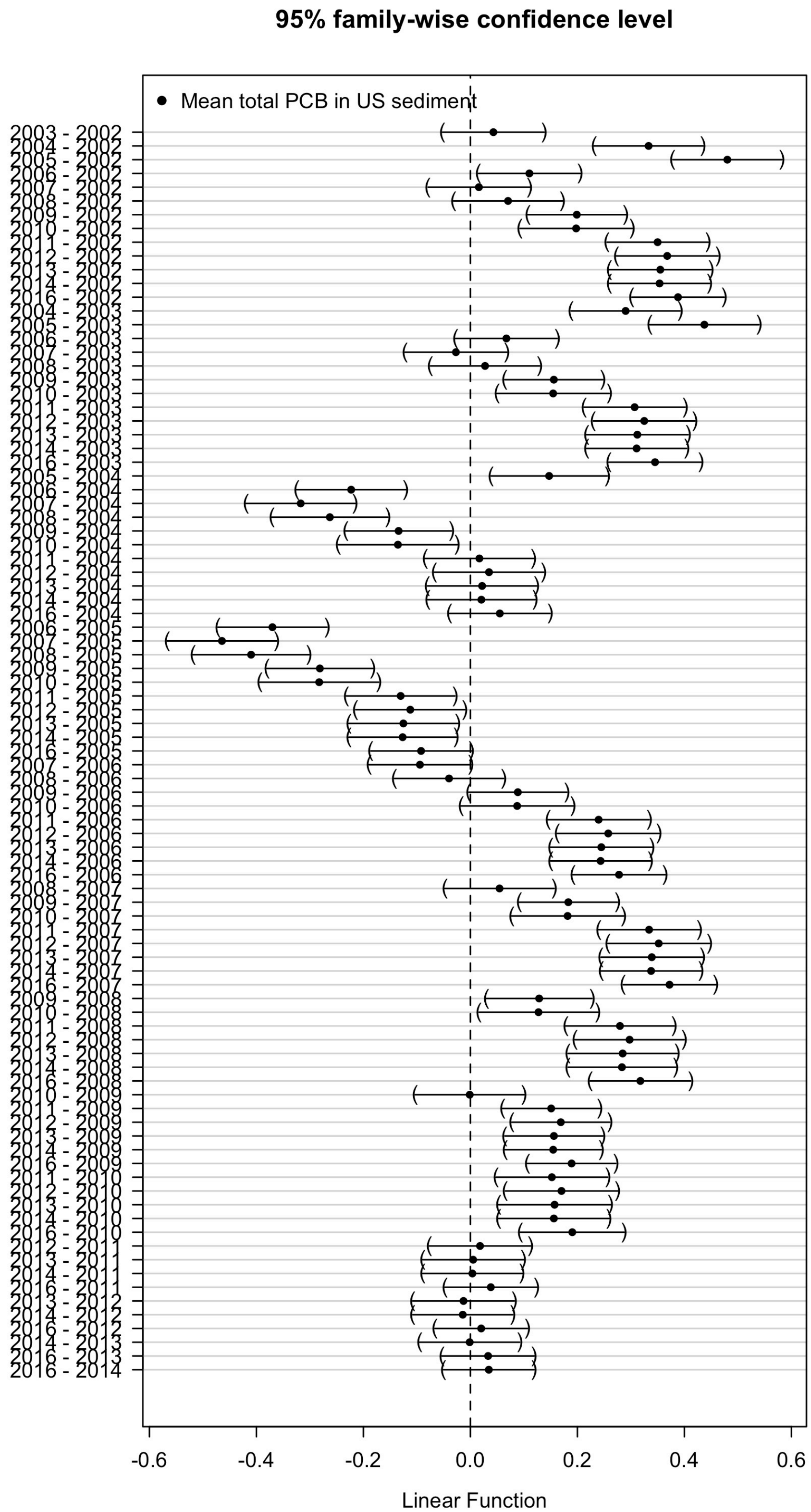


Figure B 10 Tukey Honest Significant Difference test results for comparisons of annual geometric mean ∑PCB concentration in Washington sediments.