A Risk Analysis of Legacy Pollutants: PCBs, PBDEs and New Emerging Pollutants in Salish Sea Killer Whales

by

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Abstract

Both Resident killer whales and their main food source, Chinook salmon, contain high concentrations of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs). Biopsies of killer whale and Chinook salmon samples have not measured these and other hazardous chemicals since 2009 and 2000, respectively. For this study, current samples of Resident killer whales and Chinook salmon were collected and analysed for PCBs, PBDEs, hexabromocyclododecane and other detected flame-retardants. A risk-based assessment was conducted to identify which pollutants were of greatest concern to the health of killer whales. PCBs were found to be the main contaminant of concern, although PBDEs are of growing concern due to a significant increase in concentration in killer whales over time. This study contributes to the second stage of the recovery strategy for Resident killer whales, within the Action Plan implemented by the Department of Fisheries and Oceans Canada.

Keywords: risk; killer whales; Chinook salmon; PCBs; PBDEs

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List of Acronyms

1,2-DiBB or 1,2-DBB	1,2-dibromobenzene
AhR	Aryl Hydrocarbon Receptor
BMF	Biomagnification Factors
CCAC	Canadian Council of Animal Care
CCME	Canadian Council of Ministers of the Environment
CS	Chinook Salmon
Dec 602	Dechlorane 602
DFO	Department of Fisheries and Oceans
DL	Detection Limit
DP	Dechlorane Plus
ERα	Estrogen Receptor
FEQG	Federal Environmental Quality Guideline
HBCDs or HBCDDs	Hexabromocyclododecane
HRGC	High Resolution Gas Chromatography
HRMS	High Resolution Mass Spectrometry
IC_{50}	50% Inhibition Concentration
IL-10	Interleukin 10
KW	Killer Whale
LC MS/MS	Liquid Chromatography - Tandem Mass Spectrometry
LR GC/MS	Low Resolution Gas Chromatography/Mass Spectrometry
lw	Lipid Weight
MT1	Metallothionein
PBDEs	Polybrominated Diphenyl Ethers

PBT	Pentabromotoluene
PCBs	Polychlorinated Biphenyls
QA/QC	Quality Assurance/Quality Control
RKW	Resident Killer Whale
SARA	Species at Risk Act
SD	Standard Deviation
SE	Standard Error
SQC	Sediment Quality Criteria
TBCT	Tetrabromo-o-Chlorotoluene
TEC	Toxic Equivalent Concentration
TEF	Toxicity Equivalency Factor
TRG	Tissue Residue Guideline
TRV	Toxicity Reference Value
TRα	Thyroid Hormone Receptor
US EPA	United States Environmental Protection Agency

Chapter 1.

Background

1.1. Industrial Contaminants and Killer Whales in the Salish Sea

Persistent, bioaccumulative, potentially toxic chemicals are a worldwide issue of concern as contaminants. They have the potential to impact various ecosystems and animals, especially high trophic marine mammals [1-3]. In the 1930s, polychlorinated biphenyls (PCBs), persistent organic pollutants, were manufactured in large quantities, with 97% of PCB production taking place in the United States [4, 5]. Mostly used for electronic transformers and heat resistant oils, PCBs spread globally, especially into marine environments [6]. PCBs were banned in Canada in 1977; however, products made before the ban still contain PCBs and thus PCBs continue to enter the environment today [7].

Another group of contaminants – chlorinated and brominated flame-retardants – have also started to spread across the globe, affecting various marine ecosystems [5, 8]. With the purpose of minimizing public risk resulting from fires, flame-retardants were developed in the 1970s [5]. The main chemicals involved in flame-retardants, such as polybrominated diphenyl ethers (PBDEs) and Dechlorane (or Mirex), were then used to coat upholstery or plastics [5, 9]. As further research showed that these contaminants are persistent, bioaccumulative, and possibly toxic, other chemicals, such as hexabromocyclododecane (HBCDD), Dechlorane Plus (DP) and Dechlorane 602 (Dec 602), were developed as substitutes [9-11].

High trophic marine mammals are vulnerable to the damaging effects of persistent, bioaccumulative, toxic pollutants such as PCBs and flame-retardants. Their position at the top of the aquatic food web makes such animals susceptible to the effects of these contaminants. [3, 6, 12]. Certain high trophic marine mammal populations, such as that of the Salish Sea killer whale (*Orcinus orca*), have been regarded as the most contaminated marine mammals in the world [1, 3, 5, 7].

On the Pacific Coast, there are three different ecotypes of killer whales: offshore, resident and Bigg's killer whales [7, 8, 13]. The Bigg's and offshore ecotypes are characterized as 'threatened' under the Species at Risk Act (SARA). The resident ecotype is split into two populations: the ‘threatened’ northern resident killer whale population and the ‘endangered’ southern resident killer whales. The diet of resident killer whales predominantly consists of fish, 96% of which are salmonid fish [3, 7]. Of those salmonids ingested by resident killer whales, 71.5% are Chinook salmon (*Oncorhynchus tshawytscha*) [3]. In contrast, the offshore and Bigg’s killer whales hunt marine mammals, such as seals and porpoises [7, 13]. As these animals are listed under SARA, the Critical Habitat of the Salish Sea killer whales is protected [14, 15]. Northern resident killer whales are found along the Georgia Strait, from off the coast of central Vancouver Island all the way up to southeast Alaska, whereas southern resident killer whales are found mostly south of Vancouver Island and north of Washington State. Members of the Bigg’s population have been seen off the coast of British Columbia and Washington state as often as have the resident killer whale populations [1, 8].

Not only are the Salish Sea killer whales the most contaminated marine mammals in the world, but their health is continually threatened. The Vancouver Harbour is periodically dredged of its sediment [14]. This dredged sediment, which contains various chemicals such as PCBs and PBDEs, is then disposed of both within and outside of the Critical Habitat of the Salish Sea killer whales [14]. Disposal operations take place specifically within the Johnstone Strait, located in the Northern Strait of Georgia [14]. This location resides within the Critical Habitat area of the northern resident killer whales [14]. Dredged sediment disposal operations also occur in the Southern Strait of Georgia, which is located within the Critical Habitat of the southern resident killer whales [14]. These marine mammals may be experiencing higher risks to their health due to their exposure to contaminants stemming from these ocean dredge disposal operations, as well as industrial waste and atmospheric inputs [7, 16].

Killer whales are magnificent animals that are symbolic of the Pacific west coast of British Columbia, Canada [1, 5]. These whales are also sentinel marine animals and can indicate the health of marine environments [12]. Understanding how contaminant levels of PCBs and various flame-retardants are affecting killer whale populations is important for an overall understanding of the health of the marine ecosystem.

1.2. Toxicity of Legacy Pollutants and Flame-retardants in Killer Whales

1.2.1. Effects

The characteristics of PCBs and flame-retardants have the potential to cause serious adverse effects in marine mammals [1, 12, 17]. Various captive studies and laboratory studies have investigated the health impacts of these chemicals on Salish sea killer whales [1, 10, 12, 17-19]. For example, Buckman et al. [2] found that PCBs increase the gene expression of aryl hydrocarbon receptor (AhR), estrogen receptor (ERα), thyroid hormone receptor (TRα), metallothionein (MT1) and interleukin 10 (IL-10) [2] in killer whale blubber. PCB levels that cause these physiological effects on killer whales can lead to possible PCB end points and direct toxicity reference values for killer whales [2].

Studying the chemical impacts on one species and then attributing these effects to a different but related species is subject to uncertainty. Nevertheless, it is common practice to apply known toxicological reference or effect values from one species to another [1, 7]. For example, Hickie et al. [20] used the marine mammal toxicity reference value (TRV) of 17 mg/kg determined from harbour seal (*Phoca vitulina*) blubber [21] to assess if PCBs were posing toxicological risks to the health of killer whales in the Salish sea. Comparing marine mammal TRVs to contaminant concentrations in killer whales is common practice due to the lack of direct toxicity effect studies on killer whales.

One example of a commonly-used marine mammal study that researched the toxicological effects of PCBs was a two-and-a-half-year study on captive harbour seals [21]. The study discovered that the high PCB concentrations found within the seals caused disruptions in both the immune and endocrine systems. Another study has attributed PCBs to cancer-causing effects in the California sea lion (*Zalophus californianus*) [7, 22]. PCBs produce negative health effects on marine mammals including immunotoxicity, reproductive impairment, neurological dysfunction and skeletal abnormalities [1, 23]. Based on these studies, and many others, PCBs are known to be toxic, bioaccumulative and persistent in the marine environment [1, 7]

The toxicity of PBDEs and other flame-retardants is not fully understood. PBDEs have been linked to negative health effects in weaned and juvenile grey seals (*Halichoerus grypus*), as Hall et al. found that there was a relationship between high PBDE concentrations and changes in thyroid hormone levels [10]. One study investigated the impact of HBCDD on rats and found that the contaminant had potential neurological effects with a 50% inhibition concentration (IC_{50}) value of 4μM [24]. Dechlorane (CAS# 2385-85-5), a known pesticide and flame-retardant also called Mirex, has been thoroughly investigated regarding its impacts on the environment. It was banned in 1978 in the United States, as it was found to have significant impacts on the endocrine and hepatic systems of animals [25]. This chemical was subsequently assigned a reference dose of 2.0×10^{-4} $day \cdot kg \cdot mg^{-1}$ for humans. Other Dechlorane-related chemicals, including Dec 602 (CAS# 31107-44-5), are also magnified through marine trophic food webs [9], as well as being found in sediments, soil and air [26].

There is little known about the health effects of other Dechlorane-related contaminants on marine mammals [9, 11]. Most studies concerning the Dechlorane family were conducted to give a base line of their presence in the marine environment. For example, Law et al. [11] researched the existence of over 30 alternative flame-retardants in harbour porpoise (*Phocoena phocoena*) blubber, and found that 11 of the 30 compounds were present, including PBDEs. Some chemicals, such as the alternative flame-retardant tetrabromo-o-chlorotoluene (TBCT, CAS# 39569-21-6), were present in concentrations that were reported to be less than 0.13 μg/kg wet weight.

Another brominated flame-retardant, 1,2-DiBB, is known to cause necrotic effects in BALB mice [27]; however, very little is known about this contaminant. It was found that 1,2-DiBB can exhibit acute toxicity in the liver of mice after 2 to 12 hours of exposure to either 250, 500 or 1000 mg/kg [27]. The study noted, however, that after the dosages of 1,2- DiBB were administrated, cell repair was observed towards the end of the hours of exposure.

Overall, previous research shows that PCBs and flame-retardants are present and can impact the marine environment. Further research concerning how these chemicals impact the health of killer whales and their ecological environment is needed.

1.2.2. Threshold Concentrations

Currently, there are no PCB or flame-retardant toxicity reference values (TRVs) available for killer whales due to the difficulty of directly assessing causal contaminant levels in the animal [1]. The toxicological threshold concentrations of several other marine mammals, however, have been used in past literature as comparatives to determine whether observed concentrations of contaminants in whales are causing a risk to the population's health [7, 20] (Table 1.1).

The marine mammal TRV that is most commonly used to assess if PCBs are potentially causing health risks in killer whales is derived from an immunotoxicity study conducted on harbour seals [21]. The study found that a 17 mg/kg lipid weight (lw) concentration of PCBs caused negative impacts on the immune system of harbour seals. The most recent TRV study

Table 1.1 List of toxicity reference values for marine mammals, with their associated effects and threshold values.

Contaminant	Species	Effects	Threshold Value
PCB	Harbour Seal (*Phoca vitulina*)	Immune system effects [17]	17 mg/kg_{lw}
PCB	Beluga Whale (*Delphinapterus leucas*)	Vitamin A disruption [28]	1.6 mg/kg_{lw}
PCB	Ringed Seal (*Pusa hispida*)	Hepatic gene transcript effects [29]	1.7 mg/kg_{lw}
PCB	Bottlenose Dolphin (*Tursiops truncates*)	Decreased growth rate [30]	10 mg/kg_{lw}
PCB	Harbour Seal (*Phoca vitulina*)	Endocrine and immune effects [31]	1.3 mg/kg_{lw}
PBDE	Grey Seal (*Halichoerus grypus*)	Endocrine and immune effects [10]	1.5 mg/kg_{lw}
Dechlorane (Mirex)	F344/N Rat (Reference Dose)	Liver and thyroid effects [25]	2.00×10^{-4} day•mg•kg^{-1}
HBCDD	Wistar Rat	Inhibition of dopamine [24]	4 μM (IC_{50})

concerning marine mammals involved ringed seals (*Pusa hispida*) [29 29], and concluded that a range of 1.7 mg/kg lw to 1.4 mg/kg lw concentrations of PCBs in harbour seal blubber caused hepatic gene transcription effects [29]. In addition, Mos et al. [31] concluded that a 1.3 mg/kg lw concentration of PCBs in grey seal blubber caused endocrine disruption in that animal. PCB TRVs have become lower as new studies have been conducted, suggesting that concentrations of PCBs in marine mammals may be more toxic than initially proposed.

There are few identifying TRVs for flame-retardants in marine mammals. Hall et al. [10] investigated TRVs for PBDEs in grey seals and found that concentrations of PBDEs above 1.5 mg/kg lw impacted the endocrine system.

1.2.3. Total PCBs in Killer Whales

There are many studies over the past couple of decades that have investigated concentrations of PCBs in Salish Sea killer whales [1, 2, 32, 33]. Initially, Ross et al. [1] concluded that killer whales contained such high levels of PCBs that they exceeded observed concentrations of PCBs in previous marine mammal studies. This made the Salish Sea killer whales the most contaminated marine mammals in the world.

Further research has been conducted over time on both resident and Bigg's killer whales, and it has been found that southern resident killer whales are experiencing higher concentrations of PCBs than northern resident killer whales [1, 3, 7]. This could be due to high levels of industrial and agricultural activities occurring in the foraging area of southern resident killer whales [1, 5]. Another possible reason why southern resident killer whales have higher contaminant levels than northern resident killer whales could be connected to the finding that Chinook salmon, the prey of resident killer whales, are more contaminated in the foraging area of southern resident killer whales [3].

Interestingly, Bigg's killer whales are more contaminated than resident killer whales [1, 2], possibly because the diet of Bigg's whales includes harbour seals, high trophic marine animals that are known to frequent areas of industrial activity [1, 7]. Harbour seals therefore have high concentrations of PCBs in their blubber, subsequently resulting in Bigg's killer whales ingesting high concentrations of PCBs [1, 34]. This is a plausible reason for why Bigg's killer whales are more contaminated than resident killer whales.

1.2.4. Total PBDEs in Killer Whales

Rayne et al. [8] conducted an extensive study concerning PBDEs in killer whales, wherein they compared the concentrations of PBDEs in northern resident killer whales and southern resident killer whales. It was found that concentrations of PBDEs in male southern resident killer whales (942 ± 582 ng/g lw) were greater than concentrations of PBDEs in male northern resident killer whales (203 ± 116 ng/g lw). High concentrations of PBDEs were also found in male and female Bigg's killer whales (1015 ± 605 ng/g lw and 885 ± 705 ng/g lw, respectively), likely because of the Bigg's killer whales' diet of high trophic marine mammals, as mentioned above.

One study specifically investigated concentrations of PBDEs in the blubber of southern resident killer whales [32]. Concentrations of PBDEs ranged from 2500 ng/g lw to 15,000 ng/g lw within the ecotype, and levels were highest (15,000 ng/g lw) in juvenile southern resident killer whales [32]. Concentrations of PBDEs were discovered to surpass the marine mammal TRV

(1500 ng/g lw) for PBDEs [10, 32], showing that the health of southern resident killer whales may be compromised due to PBDEs. The current concentrations of PBDEs within killer whales must be investigated to understand if the concentrations have changed over time in southern resident killer whales, as well as in other killer whale populations.

1.2.5. Influence of Age and Sex on Concentrations of PCBs and PBDEs in Killer Whales

Ross et al. [1] observed higher concentrations of PCBs in males than in females, for northern resident killer whales, southern resident killer whales and Bigg's killer whales. Males possessing higher concentrations of PCBs than females has also been known to occur in harbour seals [12]. Female killer whales having lower concentrations of PCBs than males could be related to females transferring PCBs to their calves during reproduction. The transfer of contaminants from mother to calf is also continued during the lactation period [1, 12].

Ross et al. [1] found that concentrations of PCBs in male killer whale blubber increased over time, whereas PCBs in females were found to follow a more complex relationship. For female killer whales, concentrations of PCBs in blubber decrease during the reproductive years due to maternal transfer of contaminants to their offspring [1]. As females then enter their less reproductive years (ages 40 to 45), concentrations of PCBs have been observed to increase in their blubber [1].

Conversely, in studies conducted by Krahn et al. [32, 33], female killer whales were observed to have higher concentrations of PCBs in their blubber than were male killer whales. When comparing the ages observed in the first study by Krahn et al. [32] between sexes, the males were young, with a mean age of 17, whereas the one female sampled in the study was 27 years of age. For the second study by Krahn et al. [33], the females were also older (mean age 41) than the males (mean age 16). The fact that in both studies, females were older than males may explain why the concentrations of PCBs found in females were higher than those found in males. Assessing the health risks of killer whales can be difficult due to complexities, such as age, sex and maternal transfer.

Furthermore, calving order influences the concentrations of contaminants in young killer whales. Schwacke et al. [35] conducted a contaminant risk model and observed a decrease in survivorship of calves in bottlenose dolphins (*Tursiops truncates*), which was attributed to high amounts of PCBs transferred from mothers to calves [35]. It was also concluded that second-born calves had lower concentrations of PCBs in their blubber than did first-born calves, due to the first-born calf receiving a higher load of the contaminant from the mother [33]. The time between calf births can influence the levels of persistent, bioaccumulative, toxic chemicals, since the mother can accumulate contaminants in between pregnancies [33]. Additionally, killer whale offspring have been found to have concentrations of PCBs and PBDEs in their blubber ranging from 4.0 to 17.6 times greater than those of their mother [33]. These observations concerning birth order and contaminant transfer to offspring affect calves' chances of survival and growth, potentially putting killer whale calves at a serious health risk due to PCBs. Furthermore, the roles of age and sex on the concentrations of contaminants in killer whales have only been investigated for PCBs and PBDEs.

1.2.6. Food Source

Diet is the main route of exposure for killer whales, influencing the concentrations of contaminants seen within these marine mammals [7, 20, 36]. Northern and southern resident killer whales (collectively known as resident killer whales) feed predominantly on Chinook salmon (*O. tshawytscha*) [3, 36]. Chinook salmon make up 71.5% – 96% of the diet of northern and southern resident killer whales[9, 17, 18]. The diet of resident killer whales also consists of other fish species including Pacific herring (*Clupea pallasi*), Yelloweye rockfish (*Sebastes ruberrimus*), Pacific halibut (*Hippocampus stenolepis*) and various salmonid fish, such as Chum (*O. keta)*, Coho (*O. kisutch*), Pink (*O. gorbushca*), Sockyeye (*O. nerka*) and Steelhead salmon (*O. mykiss*) [36].

Cullon et al. [3] conducted a study exploring concentrations of PCBs in Chinook salmon located in British Columbia, Canada and Washington State, USA, and found that concentrations of PCBs in Chinook salmon did not exceed the PCB USA guidelines for fish-eating wildlife. Hickie et al. [20], however, suggested that in order for 95% of the resident killer whale population to have concentrations of PCBs in their blubber below the TRV of 17 mg/kg lw, Chinook salmon would have to contain PCB concentrations of 0.008 mg/kg wet weight (ww) or less [3, 20]. Cullon et al. [3] stated that all Chinook salmon samples exceed this suggested concentration of PCBs. Overall, killer whales are exposed to persistent, bioaccumulative, toxic chemicals through their diet and this fact should be included when characterizing how both legacy and emerging pollutants affect the health of killer whales.

1.2.7. Regulatory Guidelines

In Canada, there are guidelines in place to help protect aquatic environments from the harmful effects of contaminants (Table 1.2). These guidelines include the Canadian PCB tissue residue guideline (TRG) for the prey of fish-eating wildlife (0.05 mg/kg ww [37]) and the PCB sediment quality criteria (SQC) of 0.021 mg/kg dry weight [38]. Modeling studies have been conducted to understand if these PCB guidelines protect the Salish Sea killer whales. For example, Alava et al. [7] explored a PCB food-web bioaccumulation model that investigated how effective the SQC was at protecting the resident killer whale population. An assessment of the

Table 1.2 List of Canadian guidelines for polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) for mammals that consume aquatic biota.

Contaminant	Guideline	Homolog (if applicable)	Value
PCBs	Environment Canada PCB tissue residue guideline (TRG) [37]	-	0.05 mg/kg wet weight
PCBs	Environment Canada PCB sediment quality criteria [38]	-	0.021 mg/kg dry weight
PBDEs	Canadian Federal Environmental Quality Guideline (FEQG) or PBDE Mammalian Wildlife Diet [39]	TetraBDE	44 ng/g wet weight
		PentaBDE	3 ng/g wet weight
		BDE-99	3 ng/g wet weight
		HexaBDE	4 ng/g wet weight
		HeptaBDE	64 ng/g wet weight
		OctaBDE	63 ng/g wet weight
		NonaBDE	78 ng/g wet weight
		DecaBDE	9 ng/g wet weight
HBCDDs	Canadian Federal Environmental Quality Guideline (FEQG) for HBCDD Mammalian Wildlife Diet [40]	-	40 mg/kg wet weight

relationship between concentrations of PCBs and different marine trophic levels was conducted, focusing on the concentrations of PCBs in media such as sediment, Chinook salmon, northern resident killer whales and southern resident killer whales. The model showed that PCB levels in contaminated sediment, located within the Critical Habitat of Resident killer whales, were above the PCB SQC. This and other studies also looked at Chinook salmon and found that this prey of resident killer whales exceeded the PCB TRG for wildlife consumption [7, 41]. Overall, Alava et al. [7]

showed that the current PCB SQC and TRG are not protecting the health of resident killer whale populations and thus should be revised [7].

Another study explored how long it would take for PCB levels within killer whales to reach values below marine mammal TRVs [20, 21]. The model found that by 2030, concentrations of PCBs within northern resident killer whales were predicted to be below the effects level of 17 mg/kg lw [20, 21]. For the more-contaminated southern resident killer whales, the population was projected to reach concentrations of PCBs below the TRV by 2063. Nevertheless, by 2063 both northern resident killer whales and southern resident killer whales would remain above more conservative threshold effects levels (Table 1.1) [20, 21, 31]. The parameters of the model took into account how PCB levels in killer whales were influenced by age, sex and calving order [7, 20]. Such projections show that more information is required to determine if the modeled outcome is currently occurring, and if more can be done to help these animals reach levels below the effect threshold values.

The Canadian Federal Ministry of Environment has produced Federal Environment Quality Guidelines (FEQG) for some flame-retardants. Values for the diet of mammalian wildlife have been stated for PBDEs and HBCDD, although the guidelines for PBDEs were split among several PBDE homologs [39]. For tetraBDE, the guideline was at 44 ng/g ww, whereas for pentaBDE, the mammal guideline was given at 3 ng/g ww. For hexa-, hepta-, octa- and nonaBDEs, the guidelines were 4, 64, 63 and 78 ng/g ww, respectively, and the decaBDE guideline was stated at 9 ng/g ww.

FEQG for HBCDD was also given at the value of 40 mg/kg ww for the wildlife diet of mammals [40]. Due to the recent implementation of these guidelines, no known assessment of the effectiveness of these values for killer whales has been conducted. These guidelines are, however, a helpful base line of what acceptable contaminant levels are within the environment, according to the Canadian government.

Chapter 2.

Introduction

Northern resident, southern resident and Bigg's are three populations of killer whales located in the Salish Sea. The Canadian Species at Risk Act (SARA) states that for aquatic species, it is the Canadian federal government's responsibility to protect a threatened, endangered, or extirpated animal or species in Canada [15, 42]. In 2001, the Committee on the Status of Endangered Wildlife in Canada (COSEWIC), which is responsible for classifying species that are at risk, classified the northern resident killer whale as a 'threatened' species, the southern resident killer whale as 'endangered' and the Bigg's killer whale as 'threatened' [42-44]. Reasons why these species are 'threatened' and 'endangered' include harmful and potentially toxic contaminants, fishing pressures, environmental noise, habitat disruption, increased boat traffic and climate change. These pressures can act individually and together to impact the health of the Salish Sea killer whales.

When a species is listed under the SARA, its habitat is legally protected from destruction and conservation efforts must be made for its benefit (section 33) [15, 42]. To do this, in March 2008, the *Recovery Strategy for Northern and Southern Resident Killer Whales (Orcinus orca) in Canada* was produced by the Department of Fisheries and Oceans Canada (DFO) [42, 45]. This *Recovery Strategy* outlined what had to be done in order to keep the resident killer whale population from declining, including four objectives and a list of subjects, referred to as knowledge gaps, that were to be focused on in order to better understand how to recover the resident killer whale population. The four objectives were as follows:

> Objective 1: Ensure that Resident Killer Whales have an adequate and accessible food supply to allow recovery.
>
> Objective 2: Ensure that chemical and biological pollutants do not prevent the recovery of Resident Killer Whale populations.
>
> Objective 3: Ensure that disturbance from human activities does not prevent the recovery of Resident Killer Whales.

> Objective 4: Protect critical habitat for Resident Killer Whales and identify additional areas for critical habitat designation and protection.[45]

Additionally, the list of knowledge gaps included: "1) Resident killer whale population dynamics and demographics, 2) reduced prey availability, 3) environmental contaminants, 4) physical disturbance, 5) acoustic disturbance, 6) critical habitat[45]".

Subsequently, in September 2008, the *Northern and Southern Resident Killer Whales (Orcinus orca) in Canada Critical Habitat Protection Statement* was released which, by referencing the *Recovery Strategy*, declared that resident killer whales were legally protected, as the geophysical attributes of the Critical Habitat of the Resident killer whales were stated in the *Recovery Strategy*, which was required by the SARA in Section 58 (5)[46]. The statement continued on to list human activities that affected the Critical Habitat of the Resident killer whales, such as industrial activities (construction, drilling, dredging), as well as listing the specific legislations, regulations and policies currently in place that could be used to support the protection of the Critical Habitat[46]. Other issues that could affect the features of the Critical Habitat were also mentioned, such as disturbance (i.e., whale watching), degradation of acoustic environment and prey availability. Each issue mentioned was linked with mitigation measures that directly dealt with the threats to the Critical Habitat of Resident killer whales[46].

Later in February 2009, the *Critical Habitats of the Northeast Pacific Northern and Southern Resident Populations of the Killer Whale (Orcinus orca) Order* was issued[47, 48]. This article consisted of a list of coordinates that stated the specific areas that were considered the Critical Habitat of the Resident killer whales, as well as stating that the coordinates were subject to the SARA, Section 58 (1)[48].

In 2010, a coalition of environmental groups represented by the organization Ecojustice sued the Minister of Fisheries and Oceans, and the Minister of the Environment for the failure to protect the resident killer whales' habitats[47, 49]. The environmental groups included the David Suzuki Foundation, Dogwood Initiative, Environmental Defence Canada, Greenpeace Canada, International Fund For Animal Welfare, the Raincoast Conservation Society, the Sierra Club Of Canada and the Western Canada Wilderness Committee. The court case[50] stated that the Protection Statement produced by the DFO in 2008 did not legally protect Resident killer whales, and that the DFO was required to protect species listed under the SARA (section 33)[50]. Ecojustice challenged the Protection Statement, saying that the document only gave guidelines that were voluntary, instead of giving binding laws and policies [50]. Ecojustice also challenged the Ministers about the Protection Order, arguing that the Order limited the scope of protection to only the geophysical aspects of the Resident killer whales' habitat. This excluded the need to understand and investigate the biological characteristics of the Resident killer whales' habitat, such as food availability, noise pollution and water quality [50]. Ecojustice held the Ministers accountable for these gaps in both the Resident killer whale Protection Statement and Order [47, 49, 50].

Ecojustice won the court case in 2010 [49, 50]. This was a monumental win because it demonstrated how Canadian citizens and environmental groups could hold the government accountable for the protection of species at risk using the SARA, a key instrument in Canada's ability to safeguard species at risk. It also highlighted the need for greater clarity and legislation in relation to acting to protect species at risk.

The Ministry of Department of Fisheries and Oceans appealed the court case, stating:

> that the *Fisheries Act* legally protected some aspects of the critical habitat of killer whales and could thus be resorted to as a substitute to a protection order under the SARA [51].

The appeal was dismissed, however, and the court ruled that "the *Fisheries Act* [should] not be resorted to as a substitute to a critical habitat protection order under the SARA" [51].

After the court case, adjustments and changes were made to the DFO's initial *Recovery Strategy* [43]. First, in 2011, an excerpt was included in a revised *Recovery Strategy* [52], acknowledging the Federal court ruling in 2010 and stating that amendments were being made within that current *Recovery Strategy* that would

> clarify that the attributes of critical habitat that were identified in the 2008 Recovery Strategy are in fact a part of critical habitat. Further refinement of the description of critical habitat and other potential areas for critical habitat designation will be considered through the action planning process [52].

Later, in 2014, a draft of the Action Plan was released [43]. The Action Plan included four broad strategies that were the same as the objectives laid out in the *Recovery Strategy* from both 2008 and 2010. Each broad strategy was then assigned a list of approaches and specifications that would help make the strategy successful. The strategy that is most applicable to this project was the "Broad Strategy 2. *Ensure that chemical and biological pollutants do not prevent the recovery of Resident Killer Whale populations"*[43]. There were six approaches given to Broad Strategy 2. The approaches that are most applicable, and which therefore form a foundation for this present study, are as follows:

> Approach 1: Investigate the health and reproductive capacity of Resident Killer Whales using scientific studies on free-ranging and stranded individuals, as related to chemical and biological pollution.
>
> Approach 2: Monitor the chemical and biological pollutant levels in Resident Killer Whales, their prey, and their habitat.
>
> Approach 3: Identify and prioritize the sources of key chemical and biological pollutants affecting Resident Killer Whales and their habitat. [43]

Ecojustice and various NGOs critiqued the Action Plan and comments were given [53, 54]. The overall critique and main concern about the draft was:

> the lack of separate action plans for endangered (southern) versus threatened (northern) whales, and the lack of actions needed on food supply, physical and acoustic disturbance and pollutant exposure for endangered southern Resident killer whales. [54]

In 2016, another draft was made and the critiquing parties expressed similar comments as those in 2014 [55]. This was the current situation at the time this present study was written.

This research project aims to support Broad Strategy 2, specifically Approaches 1, 2 and 3. Contaminant levels within Chinook salmon have not been investigated for PCBs since 2000 and for PBDEs since 2001. This project is a step towards understanding the current concentrations of contaminants in Chinook salmon, the main diet component of Resident killer whales.

Overall, this study will serve as an update concerning the levels of various contaminants, both within Salish Sea killer whales and in their diet. Understanding these contaminant levels and their impacts on the Salish Sea killer whales will help the DFO realize its Action Plan. By addressing the approaches for the Broad Strategy 2 in the Action Plan, this project will add to the scientific understanding of the health risks these marine mammals are facing. The outcome of this project will also supply helpful information for mitigation efforts, controlling contaminant sources, remediation efforts and assessments of disposal operations in marine waters.

2.1. Objectives

The overall aim of this study is to conduct current measurements of the concentrations of certain pollutants (i.e. PCBs, PBDEs and several chemicals that are of emerging concern) and to conduct a risk-based assessment of the effects of these pollutants on the health of resident killer whale populations, with the ultimate goal of identifying the pollutants of greatest concern to the health of the Salish Sea killer whales.

The specific objectives of this study are to:

- Collect new samples from killer whales and Chinook salmon and determine the concentrations of certain legacy pollutants (i.e. PCBs) and contaminants of emerging concern (i.e. PBDEs, HBCDD and other detected flame-retardants).
- Compare current concentrations of legacy pollutants and contaminants of emerging concern in killer whales and Chinook salmon to previously-reported concentrations.
- Evaluate the risk of measured contaminant concentrations in killer whales and Chinook salmon on the health of resident killer whale populations, based on
    - daily exposure rates
    - biomagnification factors
    - risk assessment involving measured concentrations and toxicity measurements derived from the mammalian toxicological literature.

This study aims to contribute to the second stage of the recovery strategy for resident killer whales, which is part of the Action Plan implemented by the DFO. This study helps achieve the Action Plan by investigating which chemicals are affecting the health of resident killer whales, highlighting the pollutants on which to focus. The project also measures the current pollutant levels in resident killer whales and their diet. Overall, the study will contribute information to the Action Plan's objective of "ensuring chemical biological pollutants do not prevent the recovery of resident killer whales" [43].

Chapter 3.

Methods

3.1. Sample Collection and Analysis

3.1.1. New Samples

All killer whale samples were collected under the auspices of a permit from the Animal Use Committee at Fisheries and Oceans Canada. This permit operated according to the principles of the Canadian Council of Animal Care (CCAC). Fisheries and Oceans Canada issued a scientific permit for field sampling to J. K. B. Ford [56]. All samples that were collected in the United States were done under the auspices of US ESA Permit #78-1824-01 [56].

Killer whale biopsy samples were taken in 2015 (n = 9) using a dart projector that collected skin and blubber samples [57]. The blubber samples were removed from the dart, placed into pre-cleaned aluminum foil and cryovials, and subsequently placed in liquid N_2 or at -80°C until analysis [8, 56]. Killer whale identification was also conducted for each sample using a photographic catalogue, along with one sample for a deceased whale [8, 13, 36].

New Chinook salmon samples were collected in 2014 (n = 7). Muscle tissue from the heads of adult Chinook salmon was collected at one location off southern Vancouver Island during their return from the sea and thus is not representative of the ‘local’ contaminant sources. Nevertheless, this tissue offers critical data as it belongs to the primary prey of resident killer whales [56]. Chinook salmon sub-samples and fillet samples were collected and frozen at -80°C until laboratory analysis.

Both newly-collected killer whale samples from 2015 and Chinook salmon samples from 2014 were analyzed for PCBs and emerging pollutants, including PBDEs, HBCDD, Dechlorane, Dechlorane Plus, Dec 602, pentabromotoluene (PBT),1,2-dibromobenzene (1,2-DiBB) and tetrabromo-o-chlorotoluene (TBCT; see Appendix A for a full list of contaminants analyzed). The pesticide contaminants hexachlorocyclohexane (HCH), heptachlor epoxide, alpha-endosulphan, dieldrin, endrin, beta-endosulphan, endosulphan sulphate, endrin aldehyde, endrin ketone and methoxychlor were also measured (see Appendix B), although the measurements for these contaminants were not included in analysis for this study. This is the first time HBCDD, Dechlorane, Dechlorane Plus, Dec 602, PBT,1,2-DiBB and TBCTs have been measured in Salish Sea killer whales. For the Chinook salmon samples, lipid content was determined for each sample.

For both killer whale and Chinook salmon samples, High Resolution Gas Chromatography/High Resolution Mass Spectrometry (HRGC/HRMS) was used to analyze for PCBs and PBDEs. For the analysis of HBCDD, Liquid Chromatography–Tandem Mass Spectrometry (LC MS/MS) was used, whereas for the emerging pollutants, Low Resolution Gas Chromatography/Mass Spectrometry (LR GC/MS) analyses were used. All contaminant analyses of both Chinook salmon and killer whale samples that were collected for this present study in 2014 and 2015, respectively, were conducted at Axys Analytical Ltd. in Sidney, British Columbia, Canada.

3.1.2. Compilation of Previously-collected Concentration Data

Congener specific concentrations of PCBs and PBDEs in blubber biopsies of Salish Sea killer whales were collected between 1993 and 2009, and compiled from several sources (Table 3.1). Information concerning the killer whales' respective ecotypes, along with sex and age range were included (Table 3.1). Killer whales sampled from 1993 to 2009 were located in the Salish Sea; these samples consisted of both skin and blubber tissue. Past concentrations of PCBs and PBDEs taken from Chinook salmon samples were also compiled (Table 3.2). Lipid content of the killer whale blubber samples was determined and reported for the majority of the killer whale samples. If lipid content was not reported, a value of 64.3% was assumed, following methodology from Ross et al. [1], who found that 64.3% was the average lipid content in a sample of 47 killer whales. The lipid content for all past Chinook salmon samples was also determined and included in this study. The compiled contaminant concentrations present in the killer whales and Chinook salmon from 1993 to 2009 were determined using various sample analysis techniques. Methods of sample analyses varied because of the changes in technology over time. Specific sample analyses methods are mentioned in Appendix C. These limitations of the database were noted when conducting data analyses.

All sample analyses, including those conducted using past and newly-gathered samples, followed quality assurance/quality control (QA/QC) and the United States Environmental Protection Agency (US EPA) protocols 1668 for PCBs and1614 for PBDEs [58, 59]. The Axys

Methods MLA-70 was used for the analysis of HBCDD[60] and the Axys Methods MLA-108 was used for the analysis of Dechlorane, Dechlorane Plus, Dec 602, PBT,1,2-DiBB and TBCTs[61].

Table 3.1 Number of Salish Sea killer whales sampled and analyzed based on contaminant, year and ecotype.

Contaminant	Year	Total Sample Size	Northern Resident		Southern Resident		Bigg's		Source
			Male	Female	Male	Female	Male	Female	
PCBs	1993	22	7	10	2	0	2	1	[62]
	1996	24	6	5	2	1	5	5	[62]
	2000	11	3	7	1	0	0	0	[62]
	2002	11	2	4	0	0	1	4	[62]
	2003	13	7	3	0	0	2	1	[62]
	2004	11	1	1	6 (3,3*)	0	0	3	* [32, 62]
	2006	6	0	0	5	1	0	0	[32,]
	2007	13	7	3	5*	7*	2	1	* [33, 62]
	2008	23	9	6	0	0	2	6	[62]
	2009	2	0	2	0	0	0	0	[62]
	2015	9	0	0	3	6	0	0	Present Study
PBDEs	1993	23	11	8	2	0	1	1	[8]
	1994	7	2	0	0	0	2	3	[8]
	1995	3	0	0	3	0	0	0	[8]
	1996	5	0	0	0	0	3	2	[8]
	1997	1	0	0	0	0	0	1	[8]
	2003	12	7	2	0	0	2	1	[62]
	2004	11	1	1	6 (3, 3*)	0	0	3	* [8, 62]
	2006	6	5	1	0	0	0	0	[33]
	2007	13	7	3	0	0	2	1	[62]
	2015	9	0	0	3	6	0	0	Present Study

Contaminant	Year	Total Sample Size	Northern Resident		Southern Resident		Bigg's		Source
HBCDD, Dechlorane, Dechlorane Plus, Dec 602, PBT, 1,2-DiBB, TBCT	2015	9	0	0	3	6	0	0	Present Study

Note: PCBs (polychlorinated biphenyls), PBDEs (polybrominated diphenylethers), HBCDD (hexabromocyclododecane),PBT (pentabromotoluene),1,2-DiBB (1,2-dibromobenzene) and TBCT (tetrabromo-o-chlorotoluene).

Table 3.2 Number of Chinook salmon samples analyzed based on year and contaminant.

Year	Contaminant	Sample Number	Source
2000	PCBs	12	[3]
2014	PCBs	7	Present Study
2002	PBDEs	1	Unpublished analysis from Axys
2014	PBDEs	7	Present Study
2014	HBCDD	7	Present Study
2015	Dechlorane	7	Present Study
2015	Dechlorane Plus	7	Present Study
2015	Dec 602	7	Present Study
2015	PBT	7	Present Study
2015	1,2-DiBB	7	Present Study
2015	TBCT	7	Present Study

All new samples and previously-collected concentration data were expressed on a lipid weight basis in units of $mg \cdot kg^{-1}$ lw, and were blank corrected. If the concentration of each contaminant was below the detection limit (DL) for any killer whale or Chinook salmon samples, the DL value was applied. This DL method was compared to other DL methods, including taking 50% of the DL, or not taking the DL and assuming the contaminant concentration was zero. A student t-test was conducted to analyze if there was a significant difference among the methods used to account for non-detectable concentrations. This student t-test analyzing the different DL outcomes was conducted to rule out any limitations or biases (see Appendix D).

3.2. Concentration Comparisons

3.2.1. Contaminant Concentrations Over Time

Measured contaminant concentrations in the Salish Sea killer whales and Chinook salmon for this present study were compared to previously-measured contaminant concentrations. PCBs and PBDEs were the only contaminants that could be examined over time because of the presence of historical data.

The logarithms of the concentrations of total (Σ)PCBs and ΣPBDEs in killer whales were plotted against time in years by sex and ecotype, and linear regression analyses between the logarithms of the concentrations of ΣPCBs and ΣPBDEs in sampled killer whales and time were performed. The linear regression analyses were conducted to understand if the concentrations of contaminants were changing in killer whales over time. If the calculated p-value for the linear regression analyses of the concentrations of contaminants over time was less than 0.05, the contaminants measured in killer whalers were considered to be significantly changing over time. The linear regression analyses were conducted for PCBs and PBDEs, and for each sex and ecotype. All linear regression analyses were carried out using Jmp® Software [63].

The half-lives of ΣPCBs and ΣPBDEs for different life stages of a killer whale were calculated to determine whether concentrations of PCBs and PBDEs could be expected to decline over the sampling years. The half-lives of ΣPCBs and ΣPBDEs in four different life stages of a killer whale were calculated using both of Alava et al.'s [7, 64] food-web bioaccumulation models for ΣPCBs and ΣPBDEs. Equation 3.1 shows the calculation for the summed elimination route rate constants for a killer whale. This was done for each PCB and PBDE congener.

$$\Sigma k = k_M + k_U + k_E + k_G + k_2 + k_L \qquad (3.1)$$

Where Σk represents the sum of the elimination rates in a killer whale. The metabolic transformation rate constant was represented by k_M, the urine excretion rate constant was represented by k_U, the fecal egestion rate constant was represented by k_E, the lung elimination rate constant was represented by k_2, the growth rate constant was represented by k_G, and the lactation rate constant was represented by k_L. The half-lives of ΣPCBs and ΣPBDEs were then determined to be 0.693/ Σk, and these were calculated for adult female, adult male, juvenile and calf killer whales.

The age of a killer whale could possibly affect changes observed in the contaminant concentrations present in its body over time. To understand if age was an influencing factor, a linear regression analysis exploring killer whale age versus sample year was conducted. This linear regression of age versus sample year was carried out to determine if the ages of killer whales changed significantly over the different sample periods. A p-value was calculated and if the p-value was less than 0.05, the change in age over the sample period was considered significant. This was conducted using the ages of killer whales sampled for concentrations of PCBs and PBDEs. All linear regression analyses were carried out using Jmp® Software [63].

To test if there was a change in mean age among the sample years, a one-way ANOVA test was performed between age and sample year. A p-value was calculated and if the p-value was less than 0.05, the mean age of killer whales was considered to be significantly different among the sample years. This was conducted for both PCB and PBDE contaminants, and was analyzed by splitting the samples based on ecotype and sex. ANOVA analyses were carried out using Jmp® Software [63].

To investigate the change in contaminant concentrations in Chinook salmon over time, the logarithms of concentrations of ΣPCBs and ΣPBDEs in Chinook salmon were also plotted against time.

This is the first time HBCDD, Dechlorane, Dechlorane plus, Dec 602, 1,2-DiBB, PBT and TBCT have been measured in killer whales and Chinook salmon in the Salish sea, and therefore concentrations versus time could not be investigated.

The concentrations of the emerging pollutants (HBCDD, Dechlorane, Dechlorane plus, Dec 602, 1,2-DiBB, PBT and TBCT) measured in killer whales were plotted against the ages of the sampled killer whales to investigate if the concentrations of the emerging contaminants significantly changed with age. Linear regression analyses were conducted for all the emerging contaminants using Jmp® Software [63]. P-values were calculated and if the p-value was less than 0.05, emerging contaminant concentrations were seen to significantly differ in relation to the age of killer whales.

3.2.2. ΣPBDE/ΣPCB Concentration Ratios

To compare any temporal changes in the relationship between the concentrations of PBDEs and the concentrations of PCBs in killer whales, concentration ratios were conducted as follows (Equation 3.2):

$$Concentration\ Ratio_{KW} = \frac{\Sigma PBDEs}{\Sigma PCBs} \qquad (3.2)$$

Where the concentrations of ΣPBDEs and ΣPCBs were both measured in $mg \cdot kg^{-1}$ lw. *Concentration Ratio*$_{KW}$ represents the concentration ratio in killer whales. The concentration ratios were calculated for killer whales that were sampled for both ΣPCBs and ΣPBDEs in the same sample year.

A linear regression analysis of all concentration ratios against sample years was conducted to investigate the relative change in concentrations of emerging contaminants compared to the legacy pollutant, PCBs. Linear regression analyses were conducted separately for each sex and ecotype.

The concentrations of HBCDD, Dechlorane, Dechlorane plus, Dec 602, 1,2-DiBB, PBT and TBCT were divided by the concentrations of ΣPCBs for all killer whales sampled in 2015. This was done to compare the concentrations of these 'emerging' pollutants to those of the legacy pollutant, PCBs. All concentrations were measured in $mg \cdot kg^{-1}$ lw.

Other concentration ratios were calculated where an average toxicity reference value (TRV) was incorporated into the ratio. A TRV is the exposure concentration of a contaminant that is likely to not cause harmful health effects to the animal in question and can be used to assess the health risks an animal may be experiencing [31, 65]. As such, TRVs are helpful in protecting the health of marine mammals. To calculate the average TRV for ΣPCBs, the geometric mean of all the lowest TRV concentrations of ΣPCBs was determined. The lowest TRV values for ΣPCBs used were 1.3 $mg \cdot kg^{-1}$ lw, 1.6 $mg \cdot kg^{-1}$ lw and 1.7 $mg \cdot kg^{-1}$ lw. These marine mammal TRVs were determined for harbour seals (*Phoca vitulina*), beluga whales (*Delphinapterus leucas*) and ringed seals (*Pusa hispida*), respectively. The lowest TRVs for ΣPCBs were used because they were in a similar concentration range. Additionally, these TRVs for ΣPCBs were the most current TRVs available at the time of this present study.

Once the average TRV for ΣPCBs was calculated at 1.5 mg·kg^{-1} lw, the concentrations of ΣPCBs in the sampled killer whales were divided by the average TRV for ΣPCBs (Equation 3.3).

$$\Sigma PCBs\ _{TRV} = \frac{\Sigma PCBs}{Average\ TRV\ for\ \Sigma PCBs} \qquad (3.3)$$

The concentrations of ΣPBDEs in the sampled killer whales were divided by the TRV for ΣPBDEs (1.5 mg·kg^{-1} lw; Equation 3.4).

$$\Sigma PBDEs\ _{TRV} = \frac{\Sigma PBDEs}{TRV\ for\ \Sigma PBDEs} \ (3.4)$$

These concentrations of ΣPCBs/TRV and ΣPBDEs/TRV were then plotted against the sample years and depicted on a logarithmic scale. Linear regression analyses were conducted for concentrations of ΣPCBs/TRV versus time and concentrations of ΣPBDEs/TRV versus time.

The geometric mean and standard deviation of the concentrations of ΣPCBs/TRV and ΣPBDEs/TRV versus sample year were calculated and graphed, with the y-axis plotted on a logarithmic scale. Linear regression analyses were calculated for the geometric means of both PCBs/TRV over time and PBDEs/TRV over time. All graphs were split by sex and ecotype. The outcome of these analyses is presented in Appendix I.

The ratio equations for ΣPBDEs /ΣPCBs and ΣHBCDD/ ΣPCBs were determined for the 2014 Chinook salmon samples (Equation 3.5):

$$Concentration\ Ratio_{CS} = \frac{Emerging\ Pollutant}{\Sigma PCBs} \qquad (3.5)$$

Where the emerging pollutants (mg·kg^{-1}ww) included the concentrations of either ΣPBDEs or ΣHBCDD. Concentrations of ΣPCBs were measured in mg·kg^{-1} ww. The *Concentration Ratio*$_{CS}$ represents the concentration ratios for Chinook salmon. The logarithmic conversions of ΣPBDEs /ΣPCBs and ΣHBCDD/ ΣPCBs for each Chinook salmon sample were plotted as bar graphs. The geometric mean was calculated for ΣPBDEs/ΣPCBs and ΣHBCDD/ΣPCBs, and outcomes are given in Appendix I.

The concentration ratios of PCBs and PBDEs in Chinook salmon were also calculated by incorporating Canadian wildlife consumption guidelines (Table 1.2). For PCBs, the Canadian Council of Ministers of the Environment (CCME) tissue residue guideline (TRG) for marine mammals was used; however, the TRG for PCBs was originally given in toxicity equivalents (TEQs) at a value of 0.79 ng TEQ·kg^{-1} diet ww. To obtain a CCME PCB TRG comparable to the total concentration of PCBs measured in Chinook salmon, a reworked PCB TRG was calculated (Equations 3.11 – 3.16).

The average percentages of the PCB congeners 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189 were calculated from the Chinook salmon samples collected in 2014:

$$\%\,PCB\ congener = \frac{Sum\ of\ a\ PCB\ congener}{\Sigma PCB} \qquad (3.11)$$

$$\begin{matrix} Average\ \%\ of \\ each\ PCB\ congener \end{matrix} = \frac{1}{N} \times \sum \%\ a\ PCB\ congener \qquad (3.12)$$

Where N equals the number of Chinook salmon samples. PCB congeners 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189 were chosen because they were the congeners given by the WHO 2005 [66]. The concentration of each PCB congener was then derived from the CCME PCB TRG:

$$\Sigma[C_i] = \frac{TEQ}{WHO\ TEF_i} \qquad (3.13)$$

Where TEQ was the CCME PCB TRG of 0.79 ng TEQ·kg^{-1} diet ww. TEF_i represented each WHO 2005 toxicity equivalency factor (Equation 3.13), and C_i represented the concentration of each PCB congener. Next, a new TEF was recalculated as a fraction of the guideline for the most toxic of the PCB congener, which was PCB 126, with a concentration of 7.9 ng·kg^{-1} ww (Equation 3.14):

$$TEF = \frac{TRG\ for\ PCB\ 126}{Other\ PCB\ congener\ TRG} \qquad (3.14)$$

Once the new TEF was calculated for all the PCB congeners, a toxicity equivalency concentration (TEC) for each PCB congener was determined (Equation 3.15):

$$TEC_{PCB\ congener} = \begin{matrix} Average\ \%\ of \\ each\ PCB\ congener \end{matrix} \times TEF \qquad (3.15)$$

Where the TEC was determined by multiplying the average percentage of each PCB congener in Chinook salmon samples to the newly-calculated TEF (from Equation 3.14). A new total PCB tissue residue guideline was calculated, as follows (Equation 3.16):

$$New\ PCB\ TRG = \frac{TRG\ for\ PCB\ 126}{\sum TEC_{PCB\ congener}} \qquad (3.16)$$

Where the sum of all the TEC for each PCB congener was taken and then divided by that of the most toxic PCB congener, 7.9 ng·kg^{-1} ww. The new PCB TRG was converted into mg·kg^{-1} ww with an outcome of 0.069 mg·kg^{-1} ww. The new PCB TRG based on the TEFs and TECs calculated for each PCB congener is presented in Table 3.5.

Concentrations of PBDEs in Chinook salmon samples were incorporated with the Federal Environmental Quality Guidelines (FEQG) of PBDEs for wildlife consumption, although the suggested concentrations for the PBDE FEQGs were originally stated for specific homologs (see Table 1.2). To make these guidelines more comparable to the total PBDE concentrations measured in the Chinook salmon samples, a total PBDE FEQG was calculated (Equations 3.6 – 3.10).

To do this, the average percentages of each PBDE homolog represented in the Chinook salmon samples from 2014 were first determined (Equations 3.6 and 3.7):

$$\%\ PBDE\ homolog = \frac{Sum\ of\ a\ PBDE\ Homolog}{\Sigma PBDE} \times 100\% \qquad (3.6)$$

Where both ΣPBDE homologs and ΣPBDEs were measured in mg·kg^{-1}lw. To calculate the average percentage of each PBDE homolog, the equation was:

$$\begin{matrix} Average\ \%\ of \\ each\ PBDE\ homolog \end{matrix} = \frac{1}{N} \times \sum \%\ PBDE\ homolog \qquad (3.7)$$

Where N equals the number of Chinook salmon samples. Next, a TEF was calculated for each homolog (Equation 3.8):

$$TEF = \frac{FEQG\ for\ PeBDE}{PBDE\ FEQG\ Guidelines} \qquad (3.8)$$

All the PBDE FEQGs were recalculated as a fraction of the most toxic PBDE homolog guideline, which was PeBDE, with a FEQG of 3 ng·g^{-1} ww (Equation 3.8). Once TEFs were calculated for all the FEQGs, a TEC for each PBDE homolog was calculated (Equation 3.9):

$$TEC_{PBDE\ Homolog} = \begin{matrix} Average\ \%\ of \\ each\ PBDE\ homolog \end{matrix} \times TEF \qquad (3.9)$$

Table 3.5 **A re-calculated tissue residue guideline (TRG) for polychlorinated biphenyls (PCBs) based on the toxicity equivalency concentrations (TECs) and toxicity equivalency factors (TEFs) for WHO 2005-selected PCB congeners. The average percentages of the WHO 2005 PCB congeners were based on the concentrations of PCB congeners measured in Chinook salmon samples collected in this study.**

PCB Congeners	Average Percentage*	PCB TRG (ng/kg ww)	TEF	TEC for PCB values in Salish Sea Chinook Salmon (ng/kg ww)**
PCB 77	0.03%	7.90E+03	1.00E-03	2.77E-07
PCB 81	0.00%	2.63E+03	3.00E-03	1.03E-07
PCB 105	1.5%	2.63E+04	3.00E-04	4.46E-06
PCB 114	0.09%	2.63E+04	3.00E-04	2.80E-07
PCB 118	4.00%	2.63E+04	3.00E-04	1.23E-05
PCB 123	0.07%	2.63E+04	3.00E-04	2.09E-07
PCB 126	0.01%	7.90E+00	1.00E+00	8.02E-05
PCB 156	0.38%	2.63E+04	3.00E-04	1.13E-06
PCB 157	0.05%	2.63E+04	3.00E-04	1.57E-07
PCB 167	0.15%	2.63E+04	3.00E-04	4.41E-07
PCB 169	0.01%	2.63E+01	3.00E-01	1.51E-05
PCB 189	0.03%	2.63E+04	3.00E-04	8.66E-08
				Sum of TECs (ng·kg^{-1} ww)
				1.15E-04
				*PCB TRG for Salish Sea Chinook Salmon (*mg·kg^{-1} *ww)*
				6.88E-02

*The percentage of each congener is based on Chinook salmon samples from 2014; **These new values are calculated by multiplying the average percentage of each congener by its corresponding TEF

The TEC was calculated by multiplying the average percentage of each PBDE homolog deduced from the Chinook salmon samples by the newly-calculated TEF (Equation 3.9). The sum of all the TECs for each PBDE homolog was taken and then divided by the most toxic PBDE homolog FEQG of 3 ng·g^{-1} ww, giving a new FEQG for ΣPBDEs (Equation 3.10):

$$\frac{FEQG\ for\ PeBDE}{\sum TEC_{PBDE\ Homolog}} = New\ FEQG\ for\ \Sigma PBDEs \quad (3.10)$$

The new FEQG was converted into mg·kg^{-1} ww, with an outcome of 0.00945 mg·kg^{-1} ww. The new PBDE FEQG based on the TEFs and TECs calculated for each PBDE homolog is presented in Table 3.4.

Table 3.4 The calculated Federal Environmental Quality Guideline (FEQG) for polybrominated diphenyl ethers (PBDEs) based on the toxicity equivalency concentrations (TECs) and toxicity equivalency factors (TEFs) for each PBDE homolog. The average percentages of PBDE homologs are based on the concentrations of PBDE congeners measured in Chinook salmon samples collected in this study.

PBDE Homologs	Average Percentage*	FEQG for PBDE (ng/g ww) [39]	TEF	TEC for PBDE in Salish Sea Chinook Salmon (ng/g ww)**
DiBDE	0.06%		0	0.00
TriBDE	3.01%		0	0.00
TeBDE	68.20%	44	6.82E-02	4.65E-02
PeBDE	24.20%	3	1.00E+00	2.42E-01
HxBDE	3.60%	4	7.50E-01	2.70E-02
HpBDE	0.07%	64	4.69E-02	3.33E-05
OcBDE	0.03%	63	4.76E-02	1.27E-05
NoBDE	0.24%	78	3.85E-02	9.04E-05
DeBDE	0.59%	9	3.33E-01	1.97E-03
				Sum of TECs (ng/g ww)
				3.18E-01
				*PBDE FEQG for Salish Sea Chinook Salmon (*mg·kg^{-1} *ww)*
				0.00945

*The percentage of each homolog is based on Chinook salmon samples from 2014; **These new values are calculated by multiplying the average percentage of each homolog by its corresponding TEF

Chinook salmon ratios were calculated as follows (Equation 3.11 – 3.13):

$$\Sigma PCBs_{\ TRG} = \frac{\Sigma PCBs}{TRG\ for\ \Sigma PCBs} \quad (3.11)$$

$$\Sigma PBDEs_{\ FEQG} = \frac{\Sigma PBDEs}{FEQG\ for\ \Sigma PBDEs} \qquad (3.12)$$

$$\Sigma HBCDD_{\ FEQG} = \frac{\Sigma HBCDD}{FEQG\ for\ \Sigma HBCDD} \qquad (3.13)$$

Where the TRG for PCBs was 0.069 mg·kg^{-1} ww, the FEQG for ΣPBDEs was 0.00945 mg·kg^{-1} ww and the FEQG for HBCDD was 40 mg·kg^{-1} ww for the mammalian wildlife diet of aquatic organisms. Linear regression analyses were conducted for the concentrations of $\Sigma PCBs_{TRG}$ and the concentrations of $\Sigma PBDEs_{FEQG}$ in Chinook salmon over time, to investigate if there was a change in $\Sigma PCBs_{TRG}$ and $\Sigma PBDE_{FEQG}$ in Chinook salmon over time. Concentrations of $\Sigma PCBs_{TRG}$ and $\Sigma PBDE_{FEQG}$ in individual Chinook salmon samples collected for the present study were depicted in a bar graph. The geometric means of the concentrations of $\Sigma PCBs_{TRG}$, $\Sigma PBDE_{FEQG}$ and $\Sigma HBCDD_{FEQG}$ were also plotted as bar graphs.

3.2.3. Cumulative Distributions

Contaminant concentrations in the Salish Sea killer whales were also expressed as cumulative distributions. The frequency of concentrations of PCBs and PBDEs in killer whales was compared to the average marine mammal PCB TRV (1.5 mg·kg^{-1} lw) and the marine mammal TRV for PBDEs (1.5 mg·kg^{-1} lw).

For the concentration of HBCDD in killer whales, a comparison of the cumulative distributions to the IC_{50} value of 4 μM was carried out [24]. In order to compare the IC_{50} value to HBCDD concentrations found in the Salish sea killer whales, the IC_{50} value was converted from μM to mg·kg^{-1} lw. Equations 3.14 – 3.16 show how the IC_{50} value of 4 μM was converted from μM to mg·L^{-1}. As 4 μM is equal to 4 x10^{-6}M, it was first multiplied by the molecular weight of HBCDD at 641.7 g·mol^{-1} to give an IC_{50} value in mg·L^{-1}:

$$4.0 \text{ x } 10^{-6}\ \frac{mol}{L} \times\ 641.7\frac{g}{mol} \times 1000\frac{mg}{g} = 2.567\frac{mg}{L} \qquad (3.14)$$

Equation 3.15 shows how the IC_{50} value reported in mg·L^{-1} was normalized to the protein content of the incubation medium in units of HCBDD per mg of protein. To do this, 50 μg of protein in 1 mL of incubation medium (which is equivalent to 0.02 L·mg^{-1} of protein) was used:

$$2.567\frac{mg}{L} \times 0.02\frac{L}{mg} = 0.05\ \frac{mg}{mg\ of\ protein} \qquad (3.15)$$

Equation 3.16 shows how the IC_{50} value, now in mg of HBCDD per mg of protein, was then converted to lipid weight. Protein is known to have roughly 5% of the sorptive capacity of lipids, therefore 1 g of protein is equal to 0.05 g of lipid [67]. To express the IC_{50} of HBCDD in lipid normalized concentrations, 1 g of protein was converted to 0.05 g of lipid:

$$0.05\ \frac{mg}{mg\ of\ protein} \times\ 20\frac{mg\ of\ protein}{mg\ of\ lipid} = 1\frac{mg}{mg}\ lipid\ weight \qquad (3.16)$$

The original IC_{50} value for HBCDD at 4 μM corresponds to a concentration of 1.0 x 10^{-6} mg·kg^{-1} lw. This value was plotted against the cumulative distribution for the concentrations of HBCDD measured in killer whales, in order to understand what percentage of HBCDD concentrations in killer whales were above the IC_{50} value of 1 mg·kg^{-1} lw.

On each cumulative distribution, the TRV, reference dose or IC_{50} value (both expressed in mg·kg^{-1} lw) was plotted as a vertical line, indicating killer whales that could be experiencing the representative heath risks. The percentage of killer whale samples that were above the various thresholds was then determined.

Cumulative distributions were also calculated for historical and emerging pollutants in Chinook salmon. The cumulative distributions were graphed against the log total of contaminants found in Chinook salmon, including a vertical line that represented the Canadian guidelines for the respective contaminants. The reworked CCME TRG for PCBs (0.069 mg·kg^{-1} ww), which was converted into lipid weight concentration, was graphed with the cumulative distribution for concentrations of PCBs in Chinook salmon. A 14.1% lipid content was used to convert the reworked CCME PCB guideline to a lipid weight concentration. This Chinook salmon lipid content was used from Alava et al. [7].

The reworked FEQG for PBDEs (0.00945 mg·kg^{-1} ww) was plotted on the cumulative distribution for concentrations of PBDEs in Chinook salmon. The HBCDD FEQG was presented as a vertical line for the cumulative distribution graphed for the concentrations of HBCDD in Chinook salmon. The FEQGs for both PBDEs and HBCDD were converted to a lipid weight concentration using 14.1% Chinook salmon lipid content [7]. The percentage of Chinook salmon samples that were above these guidelines was then calculated.

3.2.4. BMFs

Biomagnification factors (BMFs) were calculated for the resident killer whales based on the newly-compiled data concerning PCBs and PBDEs. The log BMF calculations were as follows:

$$logBMF = logC_{RKW} - logC_{CS} \qquad (3.17)$$

Where C_{RKW} represented the contaminant concentrations in resident killer whales and C_{CS} equalled the contaminant concentrations in Chinook salmon samples. The relative standard error (SE) was calculated as follows (Equation 3.18):

$$SE_{logBMF} = \sqrt{\left(SE_{logC_{RKW}}\right)^2 + \left(SE_{logC_{CS}}\right)^2} \qquad (3.18)$$

The anti log BMF was calculated as follows (Equation 3.19):

$$BMF = 10^{logBMF \pm SE}$$

The new BMFs for PCBs and PBDEs in resident killer whales and their prey were compared to the model-calculated BMFs from Alava et al. [7, 64]. This comparison was made to determine if the new BMF was consistent with Alava et al.'s [7] model predictions.

3.2.5. Daily Exposure Contaminant Rates

Daily exposure contaminant rates of PCBs, PBDEs, HBCDD, BDE-47, BDE-99, BDE-153 and BDE-209 in resident killer whales were calculated. The daily exposure contaminant rates were calculated as follows:

$$D = GM_{Observed\ CS} \times FR_{RKW} \quad (3.19)$$

Where the *GM* equals the geometric mean concentration of the contaminant in Chinook salmon samples ($mg \cdot kg^{-1}$ ww), FR_{RKW} equals the daily rate at which a resident killer whale feeds on Chinook salmon ($kg \cdot day^{-1}$) and *D* represents the daily exposure contaminant rate of the contaminant in resident killer whales ($mg \cdot kg^{-1}$) [3, 7]. The daily exposure contaminant rate was then calculated based on the weight of the killer whales at certain life stages (Equation 3.20):

$$D_{RKW} = \frac{D}{RKW} \quad (3.20)$$

Where D equals the daily exposure contaminant rate in the resident killer whale ($mg \cdot kg^{-1}$) and D_{RKW} is the daily exposure contaminant rate based on the weight of the resident killer whale (kg). The weights used were 5000 kg for adult male killer whales, 2700 kg for female adult killer whales and 1000 kg for juvenile killer whales [7].

The daily feeding rate of Chinook salmon by a resident killer whale (FR_{RKW}) was calculated as follows (Equations 3.21 and 3.22; Table 3.5):

$$FR_{RKW} = \%\, CS_{RKW} \times S_{RKW} \qquad (3.21)$$

$$S_{RKW} = \%S_{RKW} \times FR_{Capitve\ KW} \qquad (3.22)$$

Where $\%CS_{RKW}$ is the percentage of Chinook salmon in the total salmonid diet of resident killer whales, which was 71.5%[3]. S_{RKW} is the daily amount of salmonid consumed by a resident killer whale ($kg.day^{-1}$), FR_{RKW} equals the daily Chinook salmon feeding rate of a resident killer whale ($kg.day^{-1}$), $\%S_{RKW}$ is the percentage of the resident killer whale diet that consist of salmonids (96%[3]) and $FR_{Captive\ KW}$ represents the overall feeding rate of a captive killer whale ($kg.day^{-1}$).

The feeding rate of a captive resident killer whale ($FR_{Captive\ KW}$) was calculated as follows (Equation 3.23):

$$FR_{Capitve\ KW} = 0.277 \times (KW)^{0.663} = (3.23)$$

Where KW is the mass of the killer whale in kg. The captive killer whale feeding rate equation is borrowed from Kriete [68], who calculated a captive killer whale's food intake based on food consumption observations (Table 3.5). The main assumption made for these calculations was that the absorption efficiency of the contaminant was equal to one.

The daily exposure contaminant rate based on a resident killer whale's weight was used to assess hazard indices for Aroclor 1016, Arocloar 1254, BDE-47, BDE-99, BDE-153 and BDE-209. To do this, available reference doses ($mg \cdot kg^{-1} \cdot day^{-1}$) were compiled. The available reference doses that were used to calculate the hazard indices are shown in Table 3.6.

Hazard indices were calculated as follows (Equation 3.24):

$$HI = \frac{D}{RfD} \qquad (3.24)$$

Where D is the daily exposure contaminant rate based on killer whale weight (or dosage) in $mg \cdot kg^{-1} \cdot day^{-1}$, RfD is the reference dose ($mg \cdot kg^{-1} \cdot day^{-1}$) and HI is the hazard index. If the hazard index was greater than 1, the contaminant was determined to have the potential to cause adverse non carcinogenetic effects in the killer whale.

Table 3.5 Daily feeding rates for resident killer whales (RKWs) ingesting total salmonids and Chinook salmon specifically. All daily feeding rates are based on the life stage of a resident killer whale and are given in kg per day.

Life Stage	Body Weight (kg)	Daily Feeding Rate of Captive Whale based on Kriete [68] ($kg \cdot day^{-1}$)	Salmonids Consumed by RKW ($kg \cdot day^{-1}$)	Daily Chinook Salmon Consumption by RKW ($kg \cdot day^{-1}$)
Adult (Male)	5000	7.85E+01	7.54E+01	5.39E+01
Adult (Female)	2700	5.22E+01	5.01E+01	3.58E+01
Juvenile	1000	2.70E+01	2.59E+01	1.85E+01

Table 3.6 Reference doses for non-carcinogenic effects of contaminants relevant to this study. Reference doses are given in $mg \cdot kg^{-1} \cdot day^{-1}$. The Chemical Abstracts Service (CAS) number is also given for each contaminant.

Contaminant	CAS Number	Reference Dose for Non-Carcinogenic Effects ($mg \cdot kg^{-1} \cdot day^{-1}$)	Effects
Aroclor 1016	12674-11-2	7.00E-05	Reduced Birth Weights
Aroclor 1254	11097-69-1	2.00E-05	Immune, Dermal and Ocular Effects
BDE-47	5436-43-1	1.00E-04	Neurobehavioral effects
BDE-99	60348-60-9	1.00E-04	Neurobehavioral effects
BDE-153	68631-49-2	2.00E-04	Neurobehavioral effects
BDE-209	1163-19-5	7.00E-03	Neurobehavioral effects

A cancer risk effect for PCBs was also calculated. The available cancer potency factor used to calculate the cancer risk estimate is shown in Table 3.7.

To understand if the different life stages of resident killer whales were at risk for carcinogenic effects, a carcinogenic risk estimate was calculated (Equation 3.25):

$$CRE = D \times CR \quad (3.25)$$

Where *D* equals the daily exposure contaminant rate based on killer whale weight (dosage) in $mg \cdot kg^{-1} \cdot day^{-1}$, *CR* is the carcinogenic risk from oral exposure (per $mg \cdot kg^{-1} \cdot day^{-1}$) and *CRE* is the carcinogenic risk estimate.

Table 3.7 Excess upper bound cancer potency factor from oral exposure to polychlorinated biphenyls (PCBs), given in per $mg \cdot kg^{-1} \cdot day^{-1}$. The Chemical Abstracts Service (CAS) number is also given for the contaminant.

Contaminant	CAS Number	Cancer Potency Factor (per $mg \cdot kg^{-1} \cdot day^{-1}$)
PCBs	1336-36-3	2.00

3.3. Risk-Based List

To fully understand which contaminant of all the explored contaminants poses the greatest health risks to the Salish Sea killer whales, a risk-based list was compiled. All the results from the various risk assessment methods were considered in order to draw conclusions concerning which contaminant poses the greatest risk to resident killer whales.

Chapter 4.

Results and Discussion

4.1. Sample Collections and Analyses

The mean concentrations of ΣPCBs in killer whales for each sampled year are presented in Table 4.1. All the concentrations of ΣPCBs measured in individual killer whales, including samples collected for this present study, are shown in Appendix E.

Table 4.1 Mean concentrations of total polychlorinated biphenyls (ΣPCBs) measured in the blubber of three different killer whale ecotypes – Bigg's, northern resident (N. Res) and southern resident (S. Res) – from 1993 to 2015. Standard deviation and sample size (N) are given.

Year	Ecotype	N	Mean ΣPCB Concentration ($mg \cdot kg^{-1}$ lw)	Standard Deviation
1993	Bigg's	3	2.41E+02	5.75E+01
1993	N. Res	17	1.18E+01	9.89E+00
1993	S. Res	2	8.40E+01	1.10E+02
1996	Bigg's	10	9.78E+01	8.99E+01
1996	N. Res	11	3.29E+01	2.83E+01
1996	S. Res	4	9.12E+01	6.93E+01
2000	N. Res	10	9.55E+00	6.10E+00
2000	S. Res	1	2.48E+02	-
2002	Bigg's	5	6.73E+01	1.88E+01
2002	N. Res	6	4.29E+00	2.51E+00
2003	Bigg's	3	1.32E+02	1.37E+02
2003	N. Res	10	8.77E+00	6.48E+00
2004	Bigg's	3	1.21E+02	2.12E+01
2004	N. Res	2	4.96E+00	2.39E+00
2004	S. Res	6	2.53E+01	1.31E+01
2006	S. Res	6	7.32E+01	5.40E+01
2007	Bigg's	3	6.55E+01	2.64E+01

Year	Ecotype	N	Mean ΣPCB Concentration ($mg \cdot kg^{-1}$ lw)	Standard Deviation
2007	N. Res	11	8.97E+00	7.92E+00
2007	S. Res	12	3.80E+01	3.17E+01
2008	Bigg's	6	1.51E+02	8.97E+01
2008	N. Res	16	1.31E+01	1.34E+01
2009	N. Res	3	1.01E+01	2.86E+00
2015	S. Res	9	1.94E+01	1.66E+01

The mean concentrations of ΣPBDEs in killer whales for each sampled year are shown in Table 4.2. For concentrations of ΣPBDEs measured in individual killer whales, see Appendix E.

Table 4.2 Mean concentrations of total polybrominated diphenyl ethers (ΣPBDEs) measured in the blubber of three different killer whale ecotypes – Bigg's, northern resident (N. Res) and southern resident (S. Res) – from 1993 to 2015. Standard deviation and sample size (N) are given.

Year	Ecotype	N	Mean ΣPBDE Concentration ($mg \cdot kg^{-1}$ lw)	Standard Deviation
1993	N. Res	19	2.78E-01	6.40E-01
1993	S. Res	2	7.81E-01	6.64E-01
1993	Bigg's	2	1.38E+00	1.23E+00
1994	N. Res	2	3.45E-01	2.40E-01
1994	Bigg's	5	8.92E-01	1.14E+00
1995	S. Res	3	1.05E+00	7.85E-01
1996	Bigg's	5	9.05E-01	5.27E-01
1997	Bigg's	1	5.42E-01	-
2003	N. Res	9	5.89E-01	4.17E-01
2003	Bigg's	3	6.86E+00	9.04E+00
2004	N. Res	2	4.64E-01	3.89E-02
2004	S. Res	6	2.01E+00	8.56E-01
2004	Bigg's	3	3.21E+00	1.50E+00

Year	Ecotype	N	Mean ΣPBDE Concentration (mg·kg^{-1} lw)	Standard Deviation
2006	S. Res	6	6.90E+00	4.44E+00
2007	N. Res	10	1.04E+00	7.02E-01
2007	Bigg's	3	3.49E+00	2.05E+00
2015	S. Res	9	2.56E+00	2.12E+00

The mean concentrations of ΣHBCDD, Dechlorane, Dechlorane Plus, Dec 602, 1,2-DiBB, PBT, and TBCT in all killer whales compiled for this study are presented in Table 4.3. Once again, for all concentrations measured in individual killer whales, see Appendix E.

Table 4.3 Geometric mean concentrations of total hexabromocyclododecane (ΣHBCDD), Dechlorane, Dechlorane Plus, Dechlorane 602 (Dec 602), 1,2-dibromobenzene (1,2-DiBB), pentabromotoluene (PBT) and tetrabromo-o-chlorotoluene (TBCT) measured in killer whale samples used in the present study. Both standard deviation and sample size (N) are given.

Contaminant	Year	Ecotype	N	Geometric Mean (mol·L^{-1} lw)	Standard Deviation
HBCDD	2015	S. Res	9	2.31E-07	4.56E-01
Dechlorane (Mirex)	2015	S. Res	9	1.10E-07	2.47E-01
Dechlorane Plus	2015	S. Res	9	9.52E-08	1.54E-01
Dec 602	2015	S. Res	9	1.51E-08	4.31E-01
1,2-DiBB	2015	S. Res	9	1.41E-08	3.43E-01
PBT	2015	S. Res	9	9.06E-09	3.23E-01
TBCT	2015	S. Res	9	4.50E-09	2.07E-01

This study showed a lack of data concerning PCBs in Chinook salmon between 2000 and 2014, and concerning PBDEs between 2002 and 2014 (see Appendix E. for all Chinook salmon samples). To better understand the health implications of the diets of killer whales, more measurements of the concentrations of PCBs and PBDEs in Chinook salmon must be collected.

4.2. Concentration Comparisons

4.2.1. Linear Regression – ΣPCBs – Killer Whales

Linear regression analysis of the log-transformed concentrations of ΣPCBs measured in male killer whales in relation to time showed that there was a significant decrease in the log ΣPCB in male northern resident killer whales over time (Figure 4.1; SE = 0.00901, $p = 0.0241$). In contrast, there were no significant changes in the concentrations of log ΣPCBs over time in male southern resident killer whales (Figure 4.2) or in male Bigg's killer whales (Figure 4.3). The linear regression analyses of the log-transformed concentrations of ΣPCBs in all male killer whales in relation to time are presented in Table 4.4.

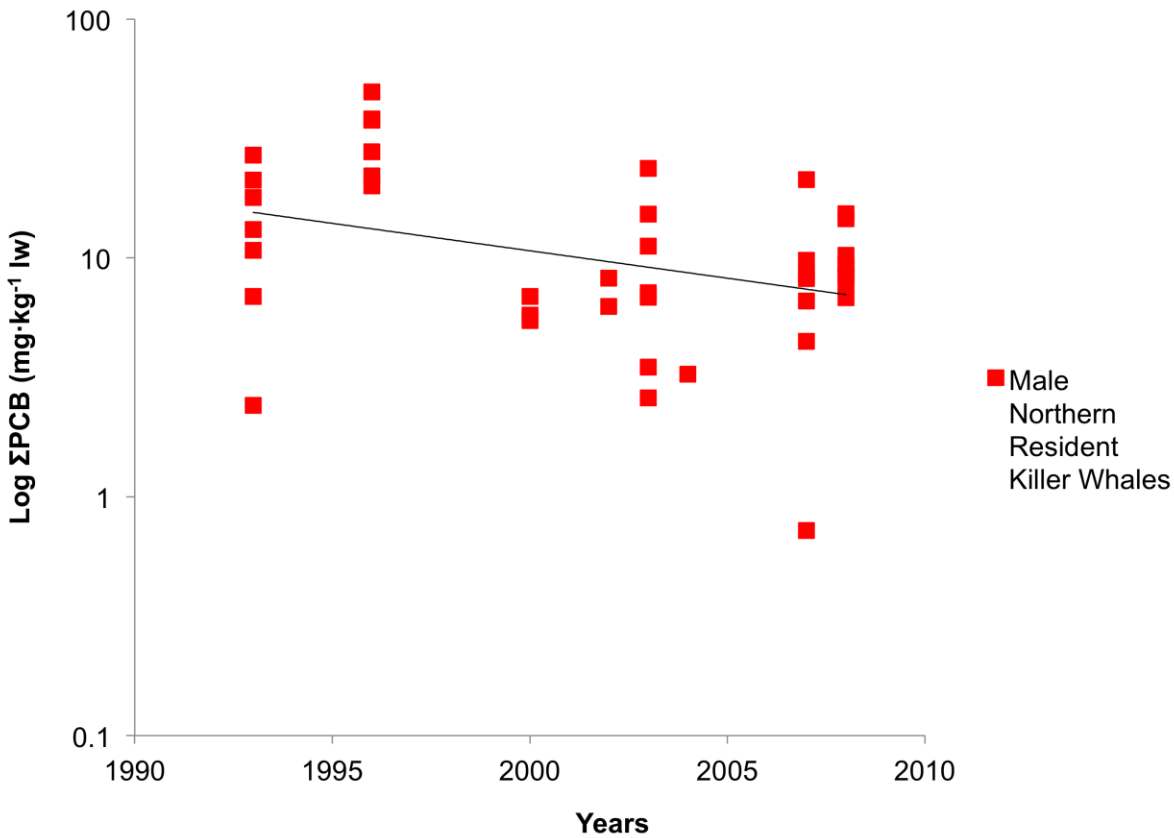

Figure 4.1 **Log concentrations of total polychlorinated biphenyls (ΣPCBs) in male northern resident killer whales in units of mg·kg^{-1} lipid weight (lw; ■) versus time in years. The solid line represents the linear regression between the log concentrations of ΣPCBs in killer whales and time.**

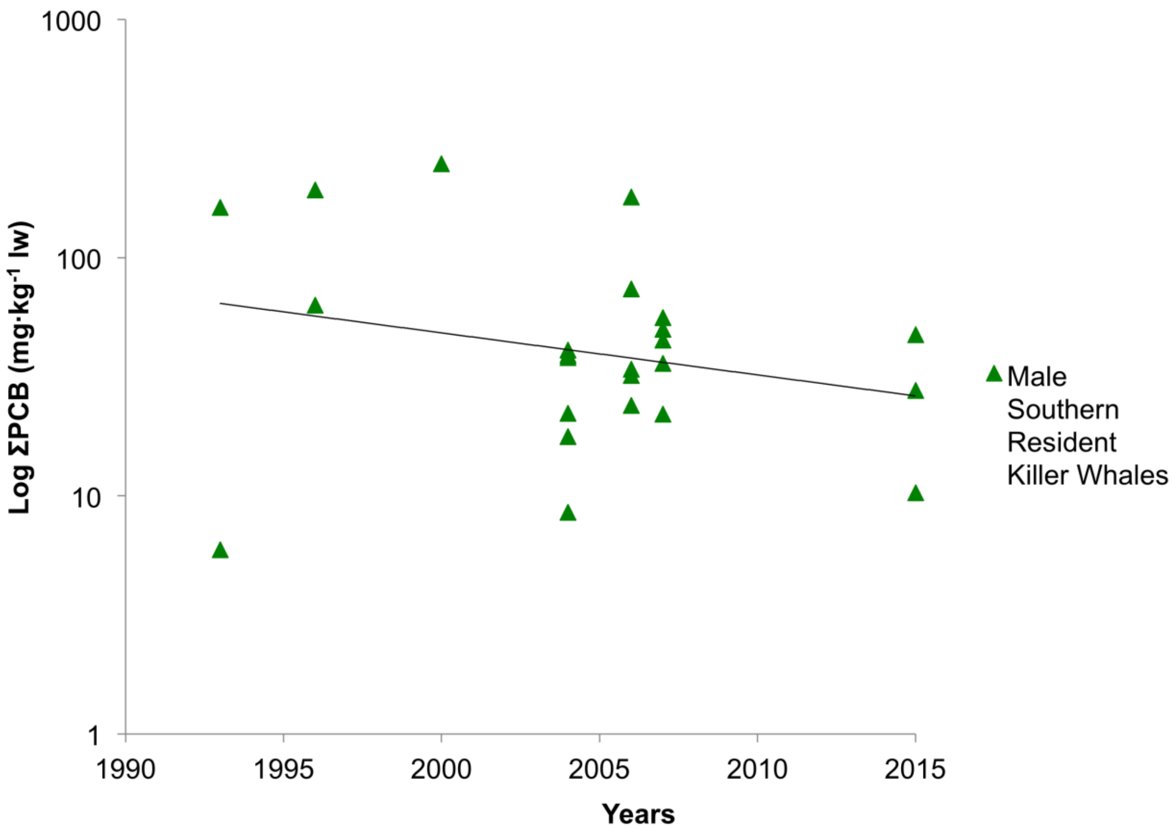

Figure 4.2 **Log concentrations of total polychlorinated biphenyls (ΣPCBs) in male southern resident killer whales in units of mg·kg^{-1} lipid weight (lw; ▲) versus time in years. The solid line represents the linear regression between the log concentrations of ΣPCBs in killer whales and time.**

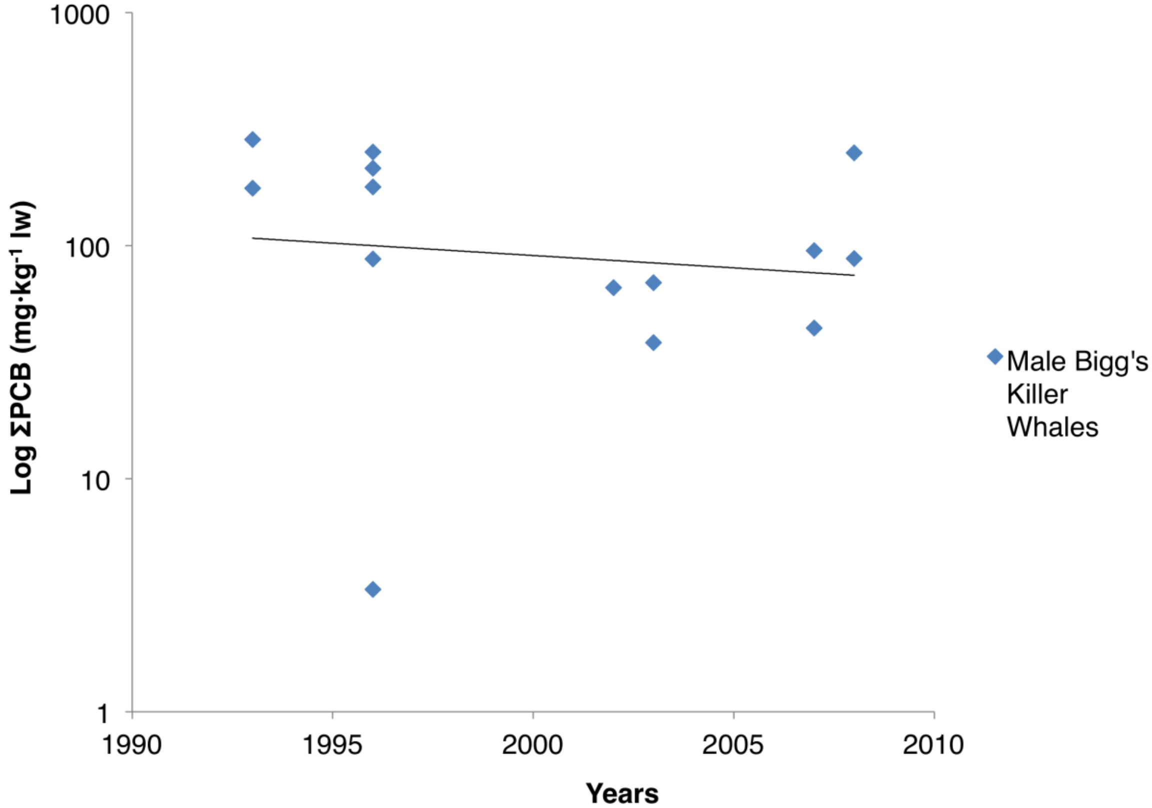

Figure 4.3 **Log concentrations of total polychlorinated biphenyls (ΣPCBs) in male Bigg's resident killer whales in units of mg·kg^{-1} lipid weight (lw; ◆) versus time in years. The solid line represents the linear regression between the log concentrations of ΣPCBs in killer whales and time.**

Table 4.4 **Outcomes of the linear regression analysis of the log-transformed concentrations of total polychlorinated biphenyls (ΣPCBs) in male killer whales over time. The analyses were split among ecotypes – northern resident (NR), southern resident (SR) and Bigg's.**

Sex	Ecotype	Linear Regression	r^2	P-value
Male	NR	Log(ΣPCB) = 43.2 - 0.0211*Year	0.115	0.0241
Male	SR	Log(ΣPCB) = 36.6 - 0.0175*Year	0.0633	0.235
Male	Bigg's	Log(ΣPCB) = 14.9 - 0.00647*Year	0.00641	0.768

Linear regression analyses of concentrations of log ΣPCBs over time in the three killer whale ecotypes showed no significant changes in the concentrations of log ΣPCBs over time in female northern resident, southern resident and Bigg's killer whales (Figures 4.4 – 4.6, respectively). The results of all linear regression analyses concerning female killer whales of each ecotype are given in Table 4.5.

Concentrations of log ΣPCBs were found to not change significantly over time in male and female southern resident and Bigg's killer whales. Female northern resident killer whales also did not exhibit significant change in concentrations of log ΣPCBs over time. This lack of significant changes in the concentrations of log ΣPCB in killer whales could be due to a slow elimination rate of PCBs in killer whales over a long period of time. Ongoing loading of PCBs into the killer whales' environment could be another reason for the lack of change in concentrations of PCBs in killer whales over time.

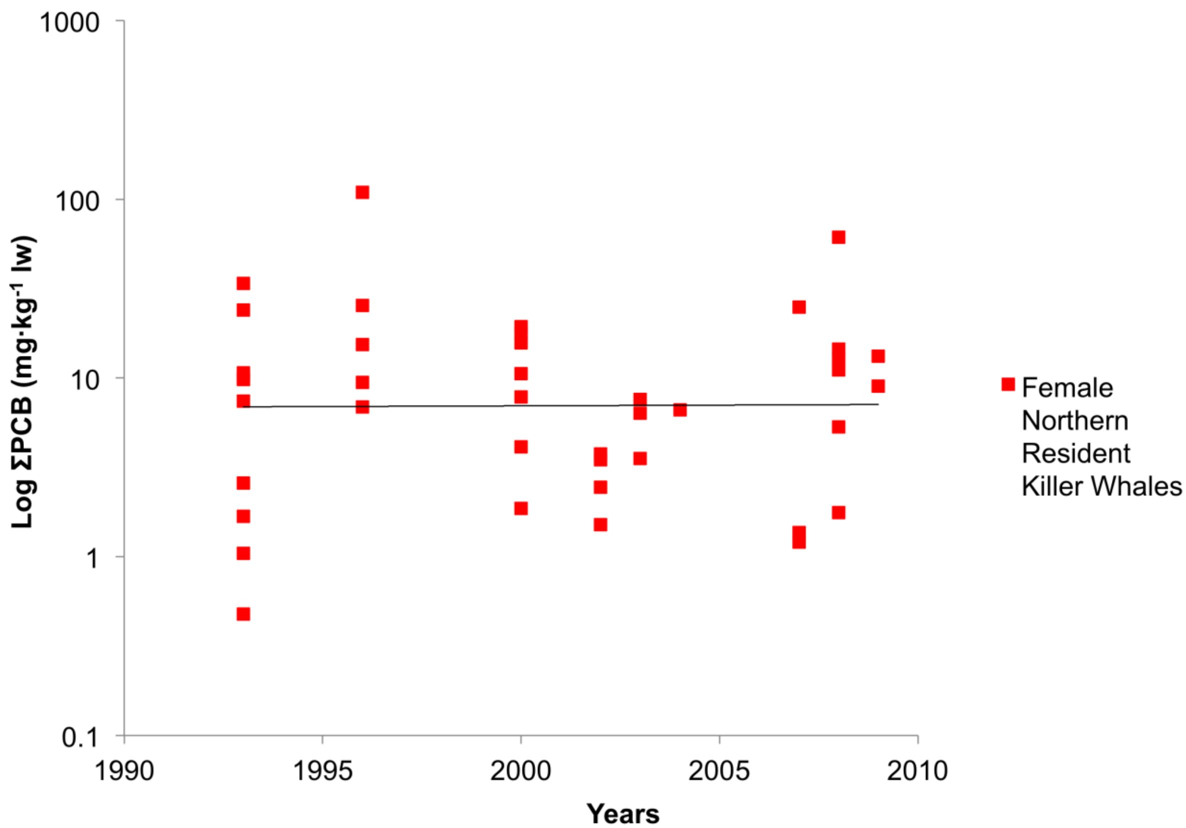

Figure 4.4 Log concentrations of total polychlorinated biphenyls (ΣPCBs) in female northern resident killer whales in units of mg·kg^{-1} lipid weight (lw; ■) versus time in years. The solid line represents the linear regression between the log concentrations of ΣPCBs in killer whales and time.

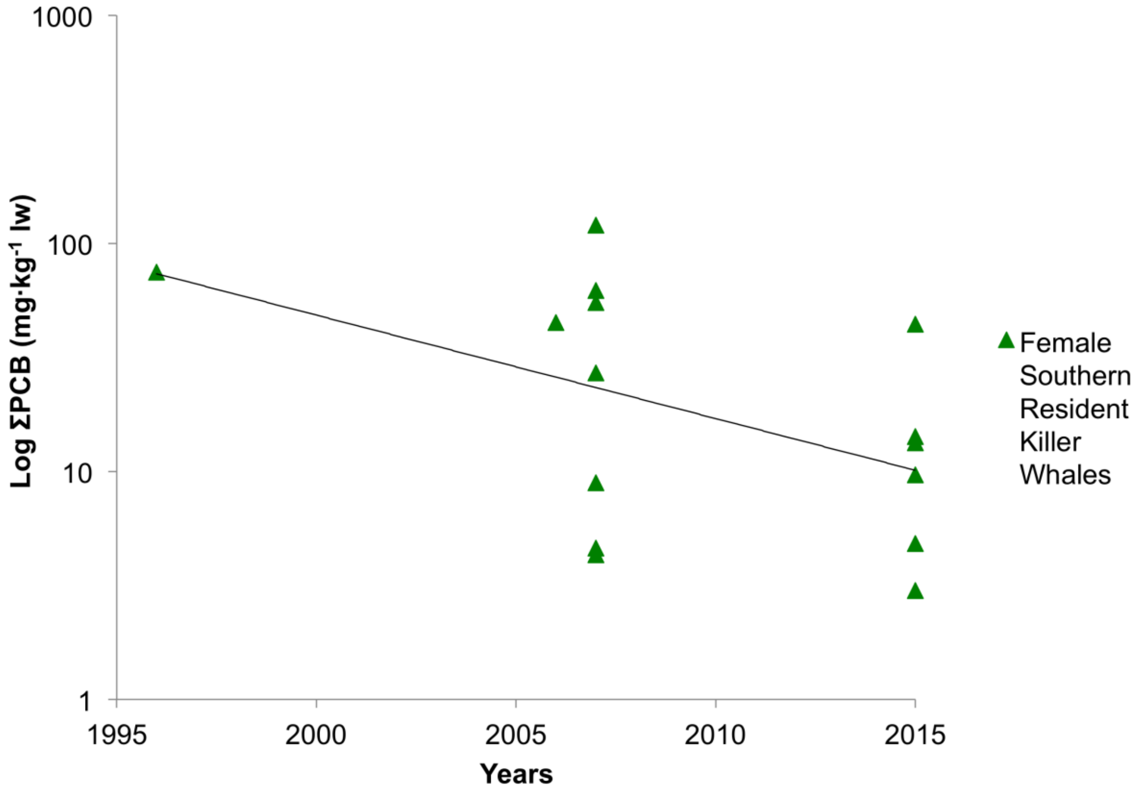

Figure 4.5 **Log concentrations of total polychlorinated biphenyls (ΣPCBs) in female southern resident killer whales in units of mg·kg^{-1} lipid weight (lw; ▲) versus time in years. The solid line represents the linear regression between the log concentrations of ΣPCBs in killer whales and time.**

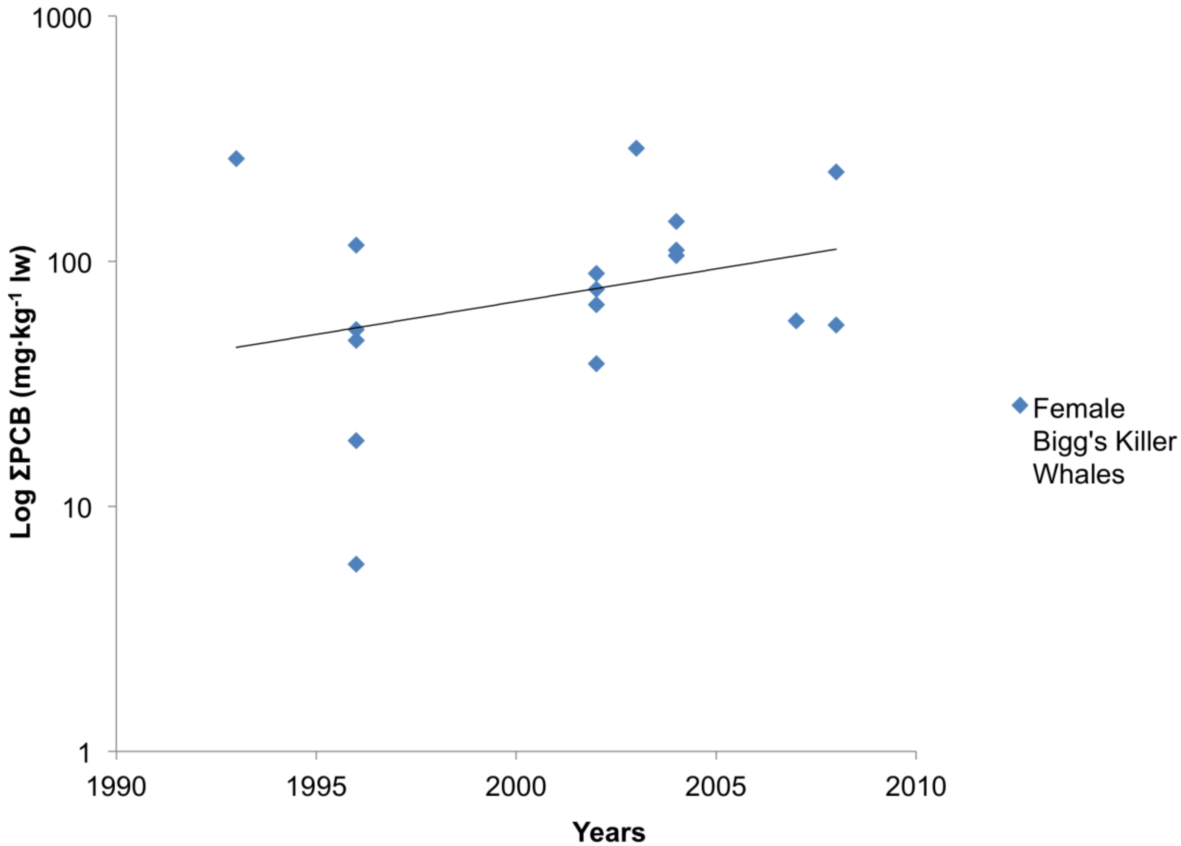

Figure 4.6 **Log concentrations of total polychlorinated biphenyls (ΣPCBs) in female Bigg's killer whales in units of mg·kg^{-1} lipid weight (lw; ◆) versus time in years. The solid line represents the linear regression between the log concentrations of ΣPCBs in killer whales and time.**

Table 4.5 **Outcomes of the linear regression analysis of the logarithmic concentrations of total polychlorinated biphenyls (ΣPCBs) in female killer whales over time. The analyses were split among ecotypes – northern resident (NR), southern resident (SR) and Bigg's.**

Sex	Ecotype	Linear Regression	N	r^2	P-value
Female	NR	Log(ΣPCB) = -1.30 + 0.00107*Year	41	0.000158	0.937
Female	SR	Log(ΣPCB) = 78.4 - 0.0384*Year	15	0.224	0.0638
Female	Bigg's	Log(ΣPCB) = -55.5 + 0.0287*Year	18	0.104	0.206

To investigate possible explanations for the lack of change in the concentrations of ΣPCBs in killer whale blubber over time, the half-lives of PCBs for four different life stages of killer whales were calculated (Table 4.6). The half-life of ΣPCBs in an adult male killer whale was determined to be 11.4 years, whereas the half-life of ΣPCBs in an adult female killer whale was 1.59 years. In contrast, ΣPCBs had a half-life of 2.84 years in juvenile killer whales and a half-life of 0.481 years in killer whale calves.

Table 4.6 Half-lives (in years) of total polychlorinated biphenyls (ΣPCBs) in killer whale blubber. The half-lives of ΣPCBs were determined for three different life stages of killer whales, with the adult stage split between sexes.

Killer Whale Life Stage	Half-life (years)
Adult Male	1.14E+01
Adult Reproductive Female	1.59E+00
Juvenile	2.84E+00
Calf	4.81E-01

The half-lives of PCBs in the different life stages of killer whales were all shorter than the 23-year time period analyzed via linear regressions of concentrations of PCBs in killer whale blubber over time (Table 4.6). A decline in concentrations of PCBs in killer whale blubber should therefore have been hypothetically observed over the 23-year time period explored in the linear regression analyses; however, no significant changes were found in concentrations of PCBs in killer whales over time. It is therefore possible that killer whales are continuously exposed to PCBs via their environment. The killer whale habitat is possibly still being polluted with PCBs.

There is a high level of variability in the sampled concentrations of PCBs in killer whales at any point in time, making it difficult to detect any changes in contaminants in killer whales over time. The monitoring program for PCBs in killer whales possesses little statistical power when detecting small changes in concentrations of contaminants in killer whales over time. Confounding factors that impact changes in contaminants in sampled killer whale blubber over time include the weight, life history exposure, calving order, sex and age of the killer whale. As such, these factors should be noted when deducing linear regression outcomes.

4.2.2. Age – PCBs – Killer Whales

It is known that as male killer whales age, the levels of their internal concentrations of PCBs can increase [33]. Females, in contrast, can offload their contaminant levels to their offspring during pregnancy and the lactation period. After the female reproduction period has ended (at approximately 40 – 45 of age), however, concentrations of PCBs in females are known to increase as they continue to age [12]. Based on these previous findings, it appears that age can impact levels of PCBs in killer whales. Accordingly, the ages of female killer whales sampled for PCBs were plotted against the years in which those killer whales were sampled for the contaminant (Figures 4.7 – 4.9). The same data were also plotted for male killer whales (Figures 4.10 – 4.12).

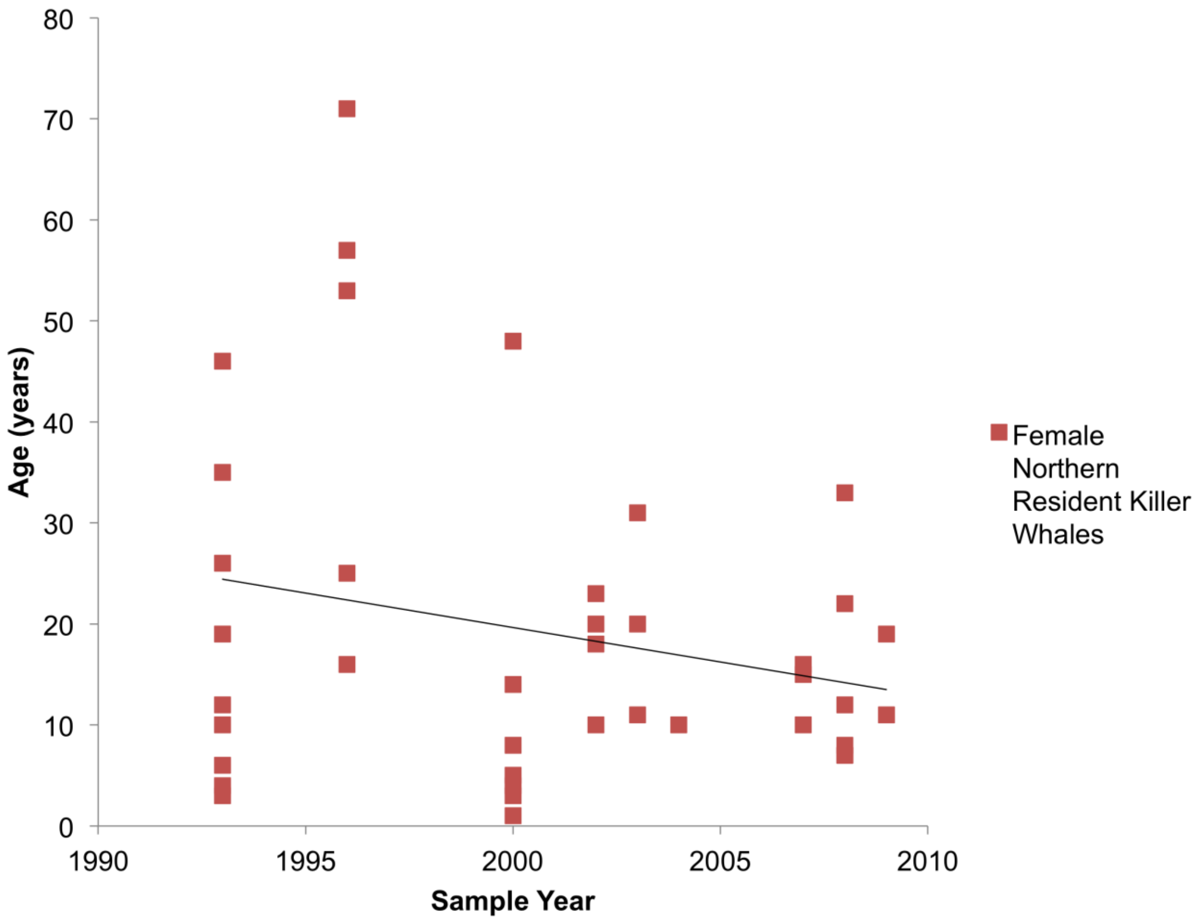

Figure 4.7 **The age of female northern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year each killer whale was sampled. The linear regression line shows the linear relationship between age and sample year.**

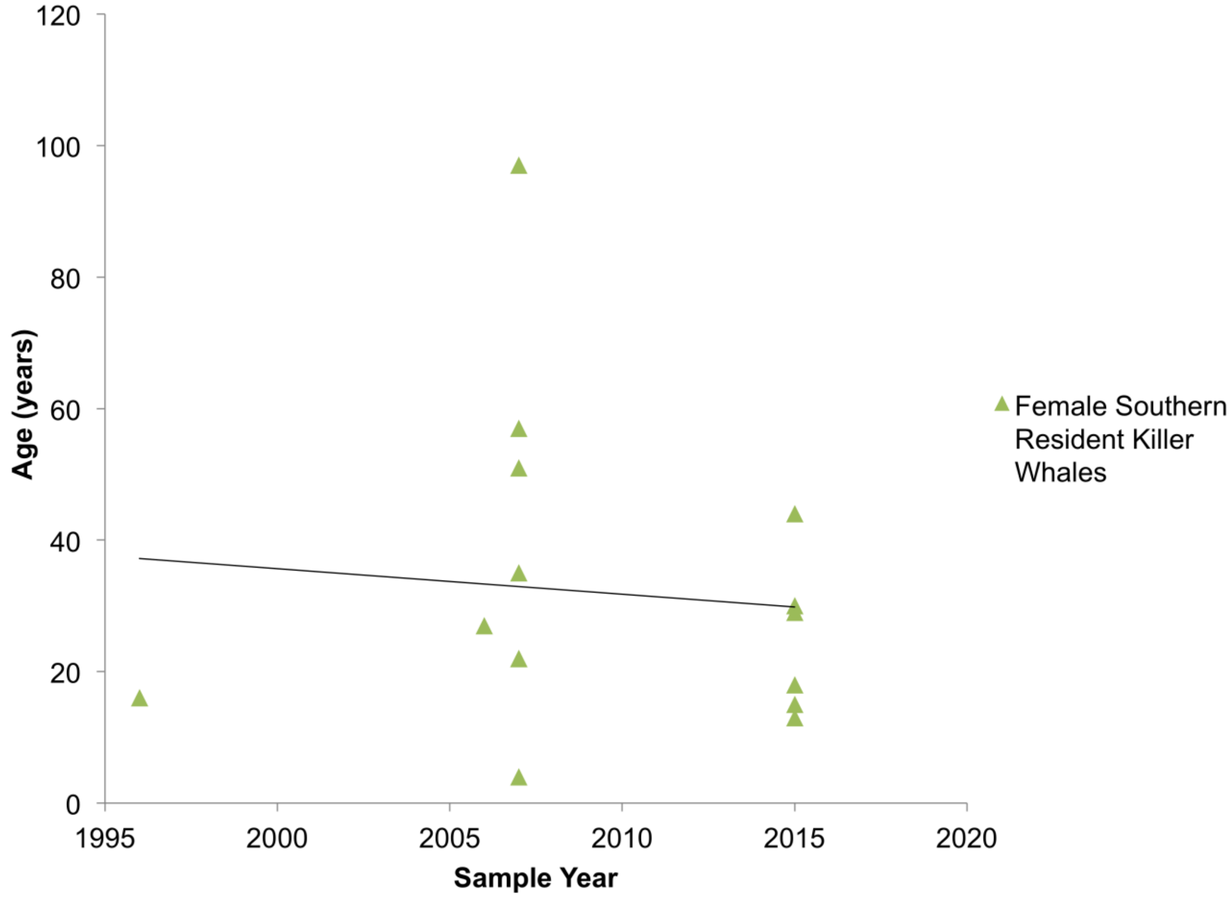

Figure 4.8 **The age of female southern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year each killer whale was sampled. The linear regression line shows the linear relationship between age and sample year.**

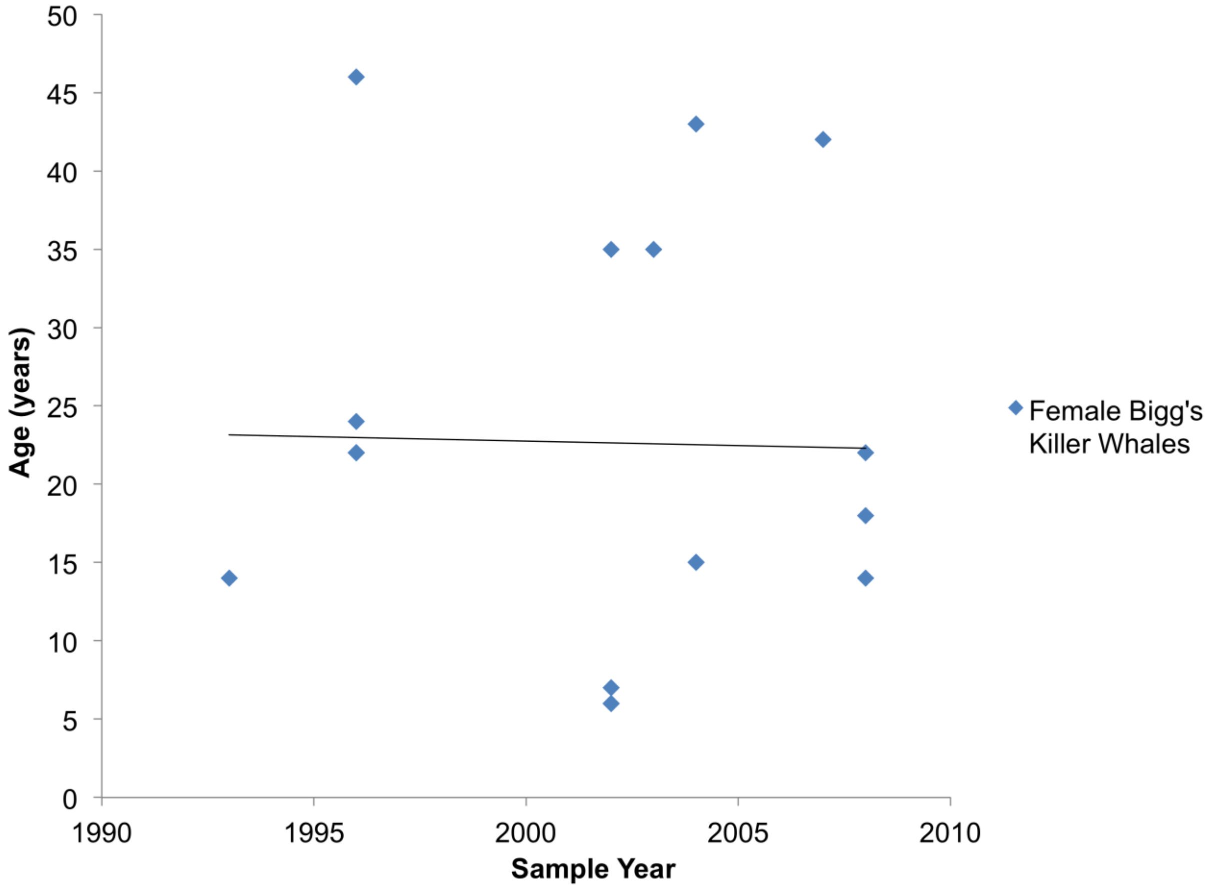

Figure 4.9 **The age of female Bigg’s killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year each killer whale was sampled. The linear regression line shows the linear relationship between age and sample year.**

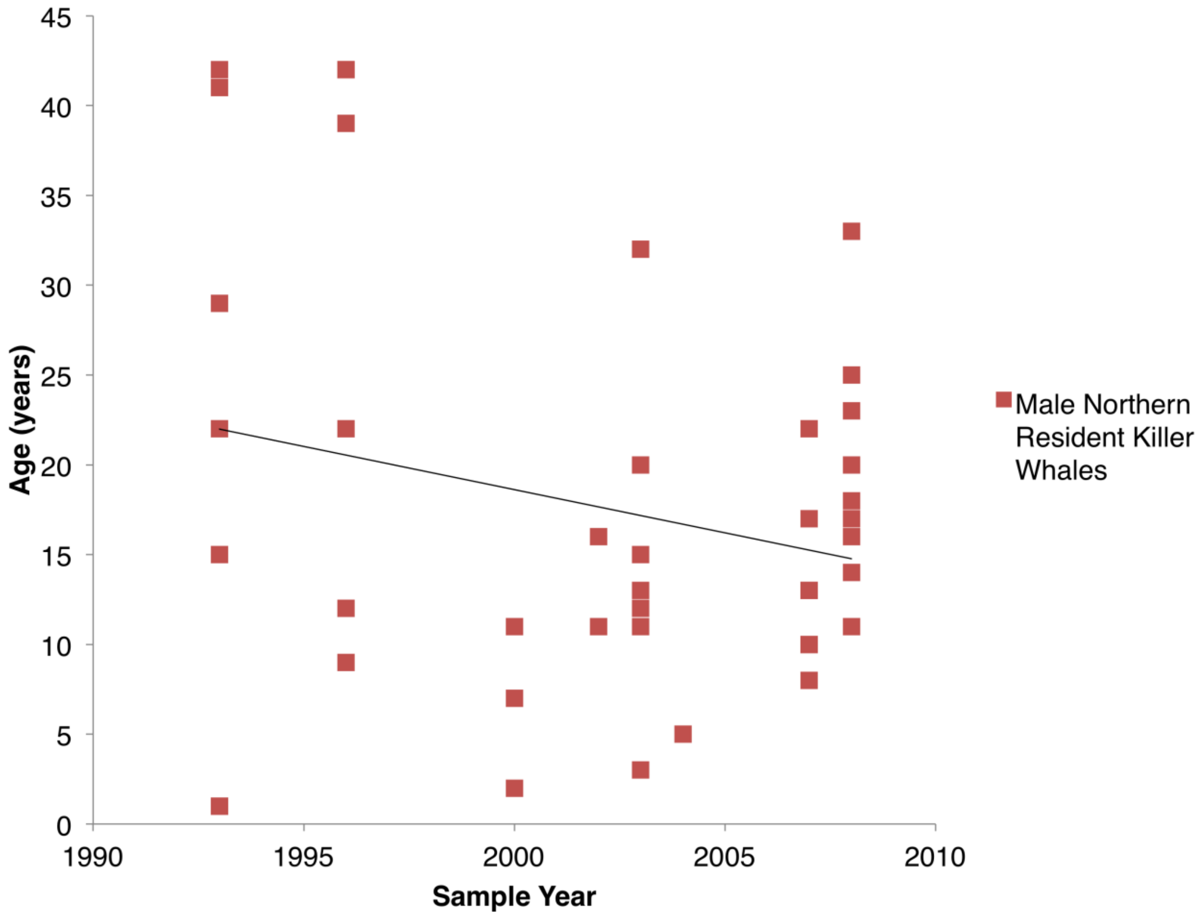

Figure 4.10 **The age of male northern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year each killer whale was sampled. The linear regression line shows the linear relationship between age and sample year.**

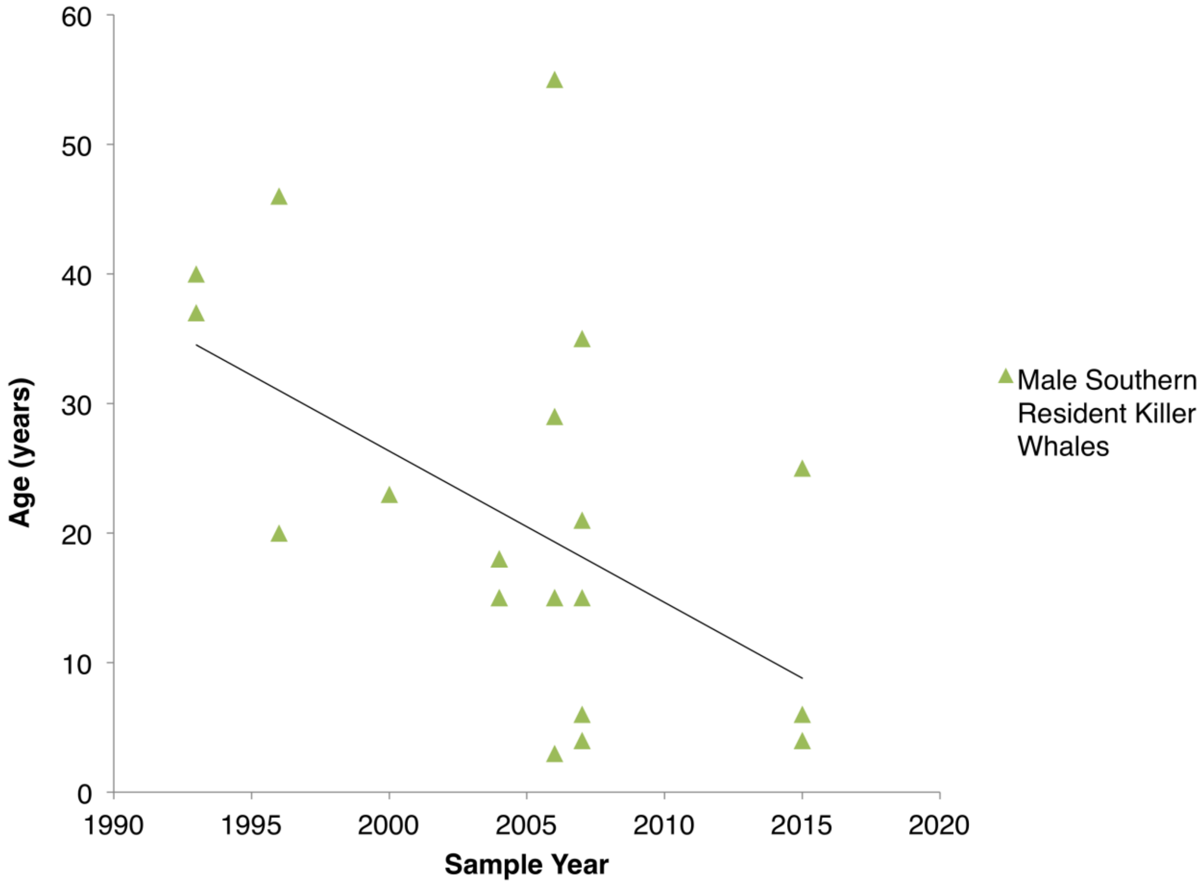

Figure 4.11 **The age of male southern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year each killer whale was sampled. The linear regression line shows the linear relationship between age and sample year.**

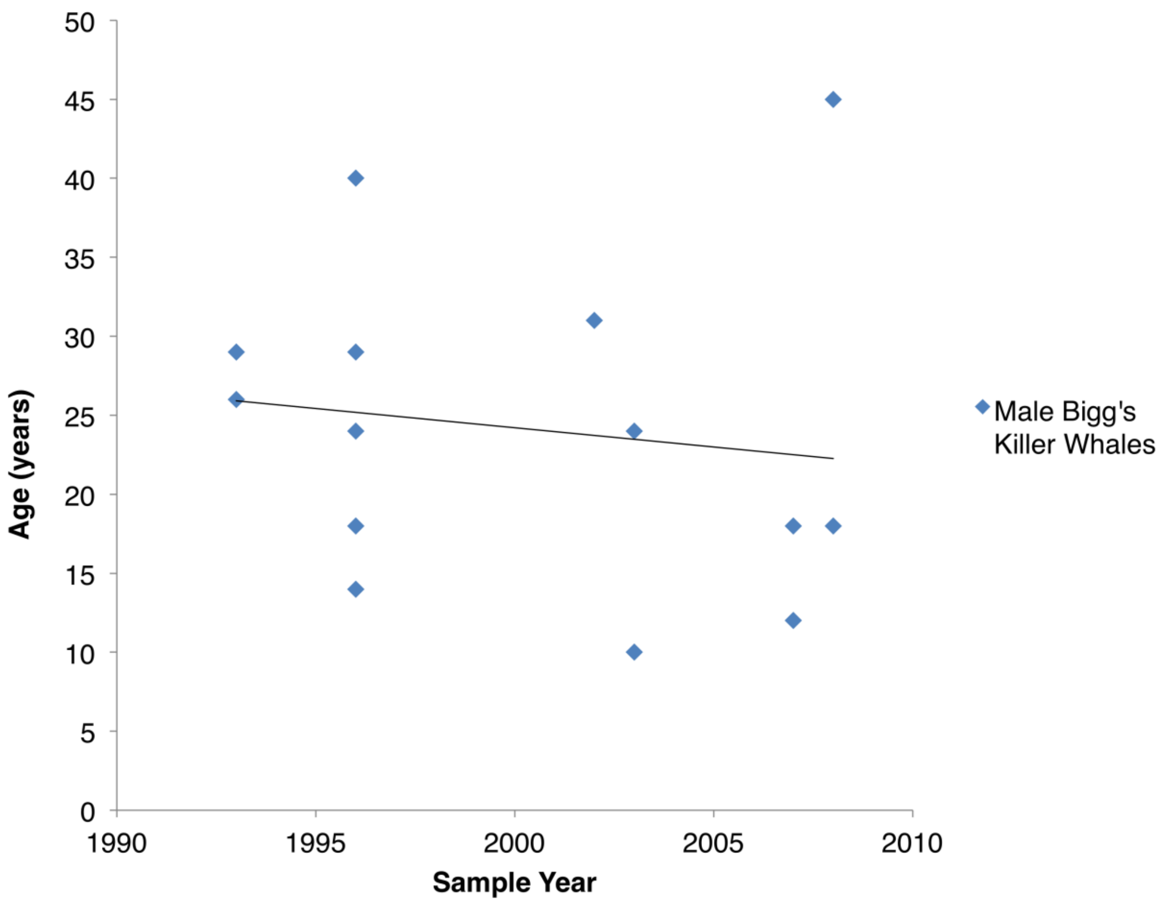

Figure 4.12 The age of male Bigg's killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year each killer whale was sampled. The linear regression line shows the linear relationship between age and sample year.

The results of the linear regression analysis of age versus time for both male and female killer whales sampled for PCBs are shown in Table 4.7. The linear regression found that the ages of all female killer whales, and male northern resident and Bigg's killer whales, did not significantly change over the years in which PCBs were measured. The age of male southern resident killer whales, however, significantly decreased over the sampled years (SE = 0.422, p = 0.0112).

Table 4.7 **Results of a linear regression comparing the age of killer whales to the years in which the killer whales were sampled for polychlorinated biphenyls (PCBs). P-values < 0.05 are considered to indicate a significant change in the age of killer whales over the sample period. Results are split by sex and by ecotype – northern resident (NR), southern resident (SR) and Bigg's.**

Sex	Ecotype	Linear-Regression	r^2	P-value
Females	NR	Age = 1409 - 0.695*Year	0.0611	0.119
Females	SR	Age = 813 - 0.389*Year	0.00846	0.745
Females	Bigg's	Age = 137 - 0.0572*Year	0.000481	0.931
Males	NR	Age = 982 - 0.482*Year	0.0559	0.132
Males	SR	Age = 2364 - 1.169*Year	0.259	0.0112
Males	Bigg's	Age = 512 - 0.244*Year	0.0187	0.641

The results of ANOVAs investigating differences in mean age among sample years are shown in Figures 4.13 – 4.18. ANOVAs were conducted separately for female and male killer whales based on ecotype.

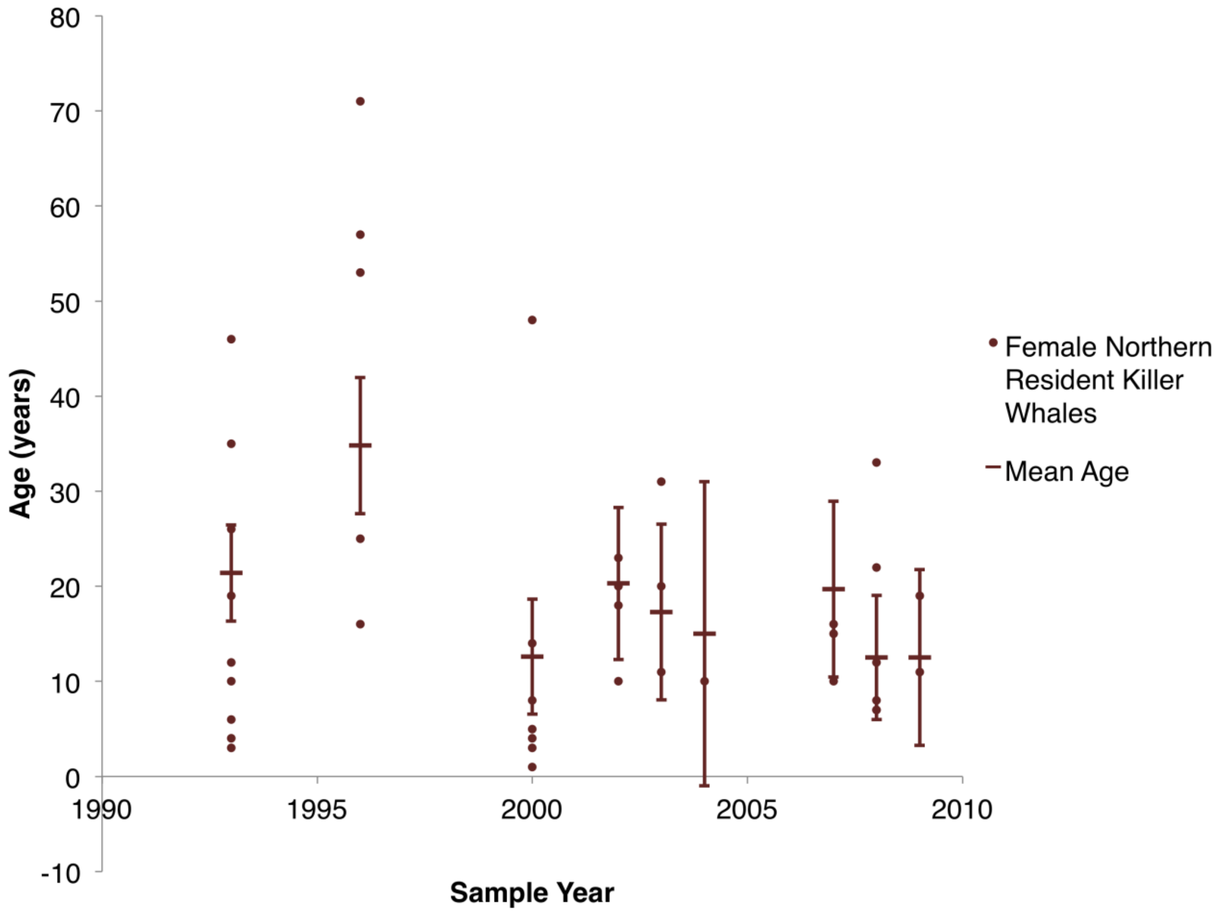

Figure 4.13 **The age of female northern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of the killer whales for that sample year.**

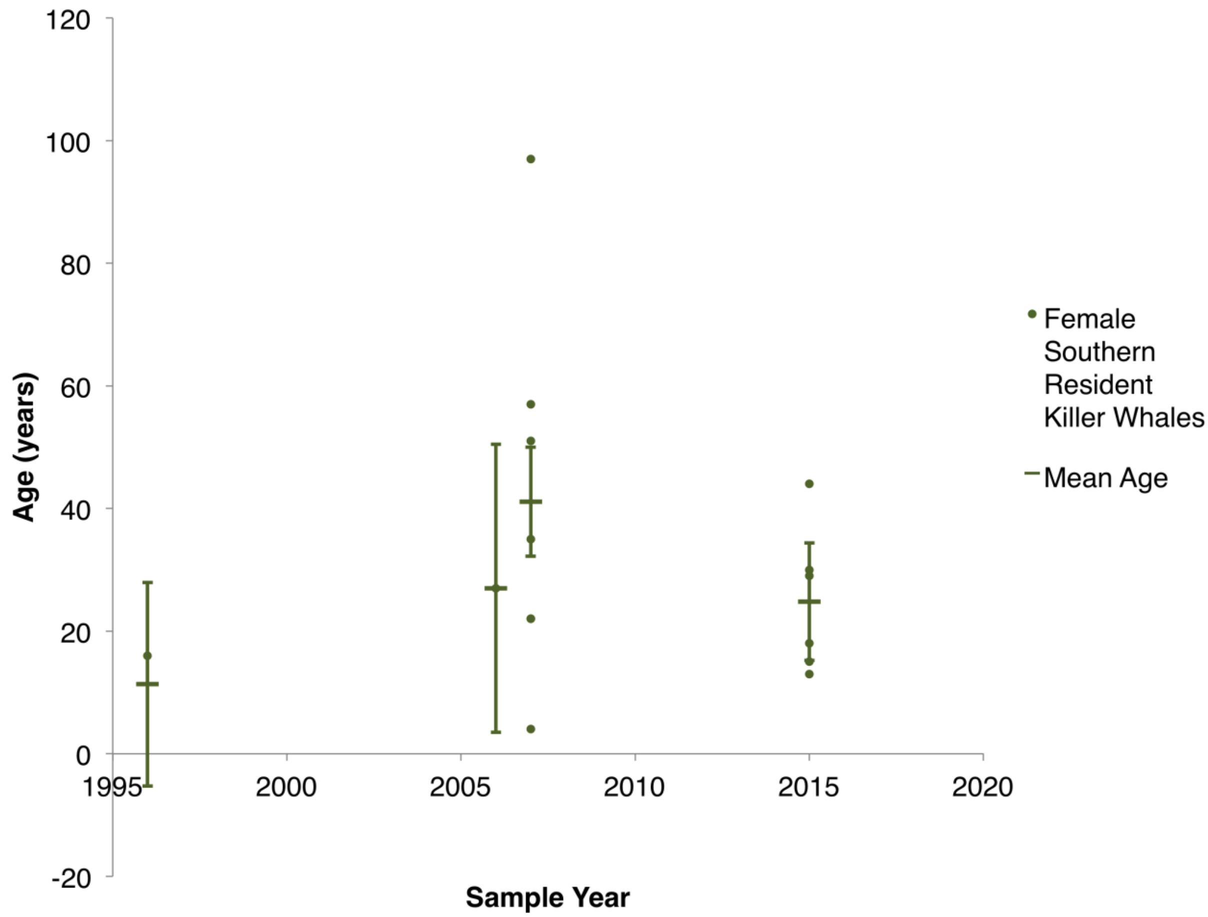

Figure 4.14 **The age of female southern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of the killer whales for that sample year.**

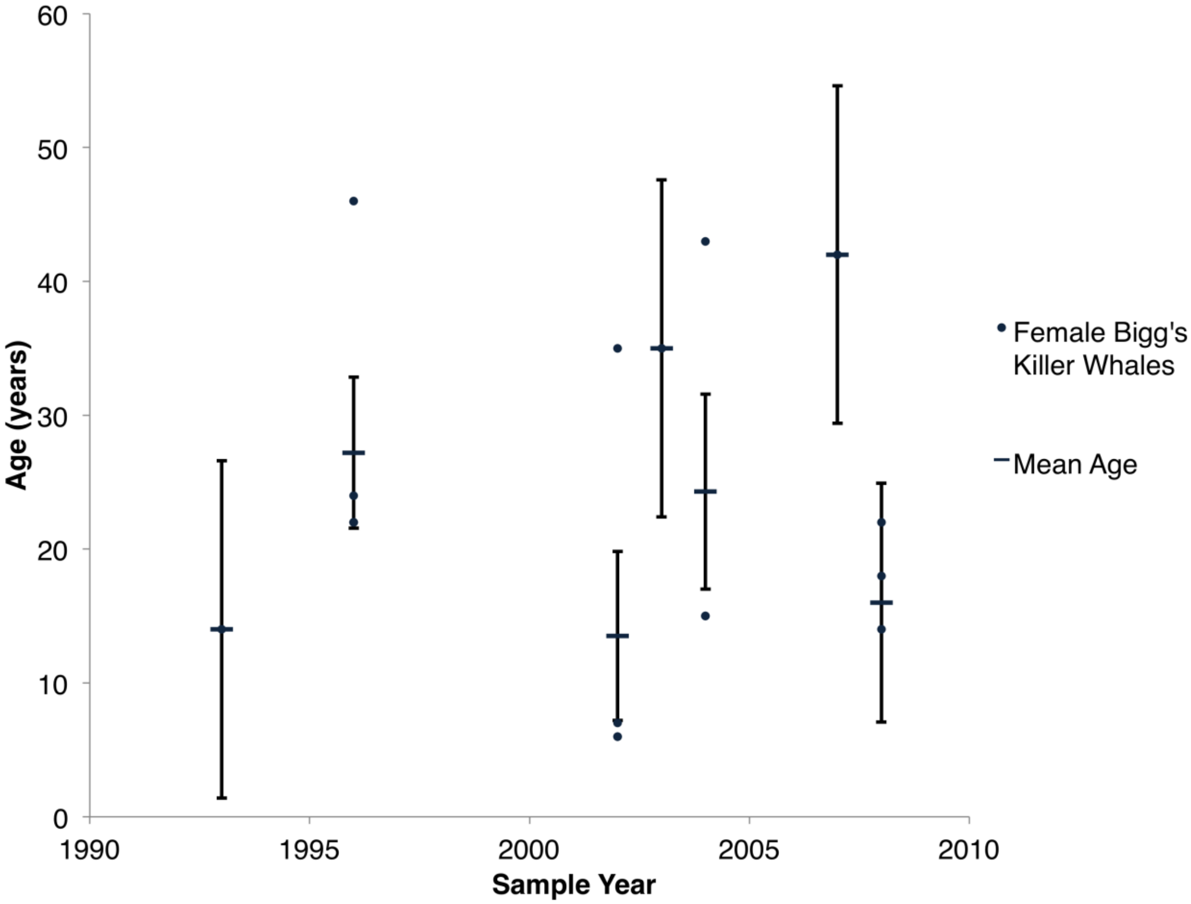

Figure 4.15 **The age of female Bigg's killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of the killer whales for that sample year.**

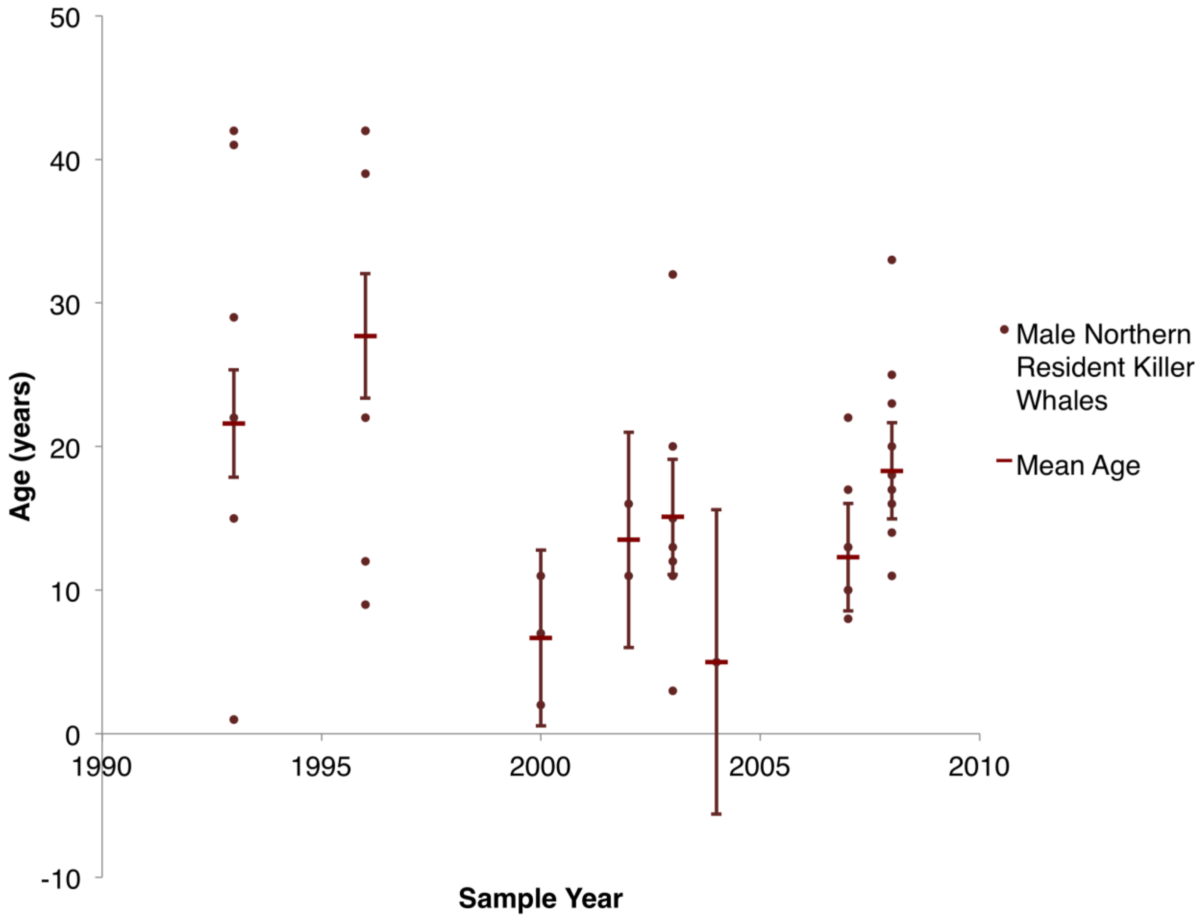

Figure 4.16 **The age of male northern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of the killer whales for that sample year.**

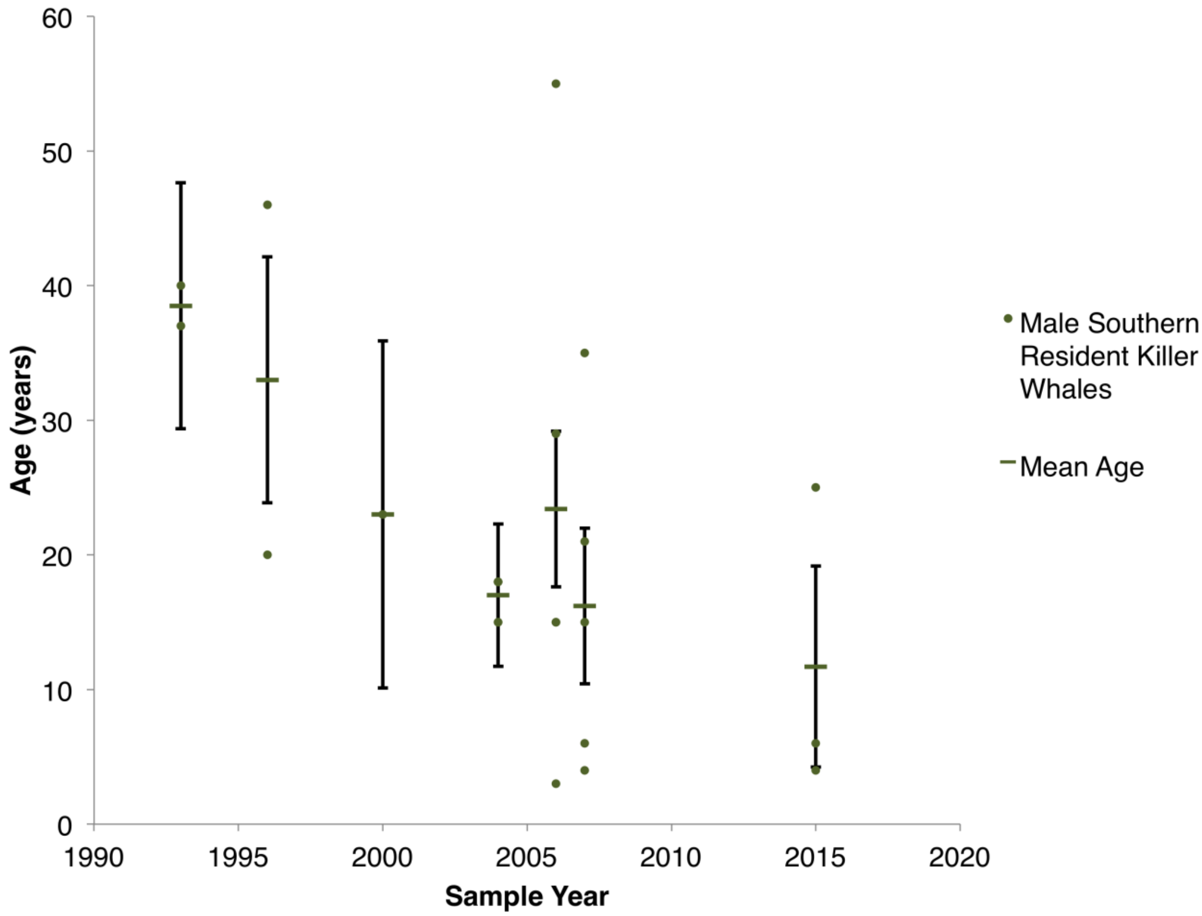

Figure 4.17 **The age of male southern resident killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of the killer whales for that sample year.**

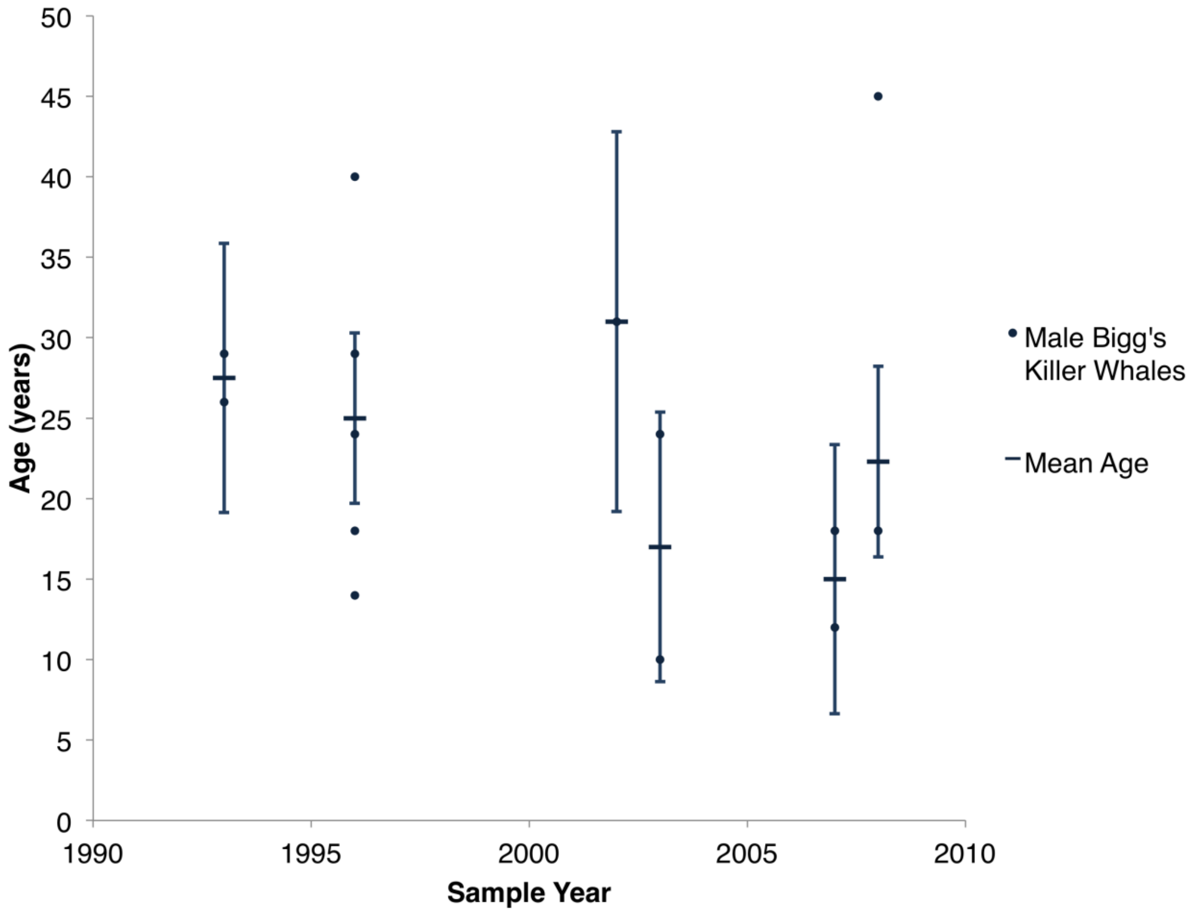

Figure 4.18 **The age of male Bigg's killer whales sampled for polychlorinated biphenyls (PCBs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of the killer whales for that sample year.**

Results of the ANOVAs conducted separately for both male and female killer whale sampled for PCBs, split by ecotype, are shown in Table 4.8. The ANOVAs indicated that the mean age of both male and female killer whales did not significantly change among the years in which PCBs were sampled, regardless of ecotype (see Appendix G for mean results for all ANOVAs).

Table 4.8 **Results of ANOVAs comparing the mean age of killer whales to the years in which the killer whales were sampled for polychlorinated biphenyls (PCBs). ANOVAs were conducted separately by sex and by ecotype – Bigg's, southern resident (SR) and northern resident (NR). P-values < 0.05 are considered to indicate a significant change in the mean age of killer whales over time.**

Sex	Ecotype	F Ratio	P-value
Females	Bigg's	1.19	0.384
Females	SR	0.566	0.648
Females	NR	1.00	0.452
Males	NR	1.98	0.084
Males	SR	1.40	0.270
Males	Bigg's	0.465	0.794

The age of killer whales over time was explored to determine if age was impacting the change in concentrations of PCBs in killer whales over time. The age of killer whales did not significantly change over time, except in the case of male southern killer whales, in which age significantly decreased over time, showing that younger animals were sampled more recently. There were no significant changes, however, in the concentrations of PCBs in male southern resident killer whales over time. Because male southern resident killer whales did not exhibit changes in concentrations of PCBs over time, it was concluded that the significant change in age in this sex and ecotype did not affect the data set.

The mean age of killer whales was also investigated and no change in mean age was found among the different sample years. Similarly, Rayne et al.[8] investigated if age varied among different sample years and also found that age was not related to the concentrations observed. The ANOVA results, along with the lack of significant changes in the concentrations of PCBs over time in male southern resident killer whales, allowed this study to conclude that age was not a sampling artefact for male southern resident killer whales. Age not being considered a sampling artefact for concentrations of PCBs in killer whales over time strengthens the findings that concentrations of PCBs are not changing in killer whales over time. This could be due to inputs of PCBs into the killer whale environment. Overall, these findings indicate that PCBs could still be considered to be a threat to killer whales.

4.2.3. Linear Regression – ΣPBDEs – Killer Whales

Linear regression analysis of the concentrations of the log-transformed concentrations of ΣPBDEs measured in male killer whales over time indicated that log ΣPBDEs in northern resident male killer whales significantly increased over time (Figure 4.19; SE = 0.0112, $p \leq 0.000100$). In contrast, male southern resident and Bigg's killer whales showed no significant changes in log concentration of ΣPBDEs over time (Figures 4.20 and 4.21, respectively). Results of the linear regression of the log concentrations of ΣPBDEs in male killer whales over time are given in Table 4.9.

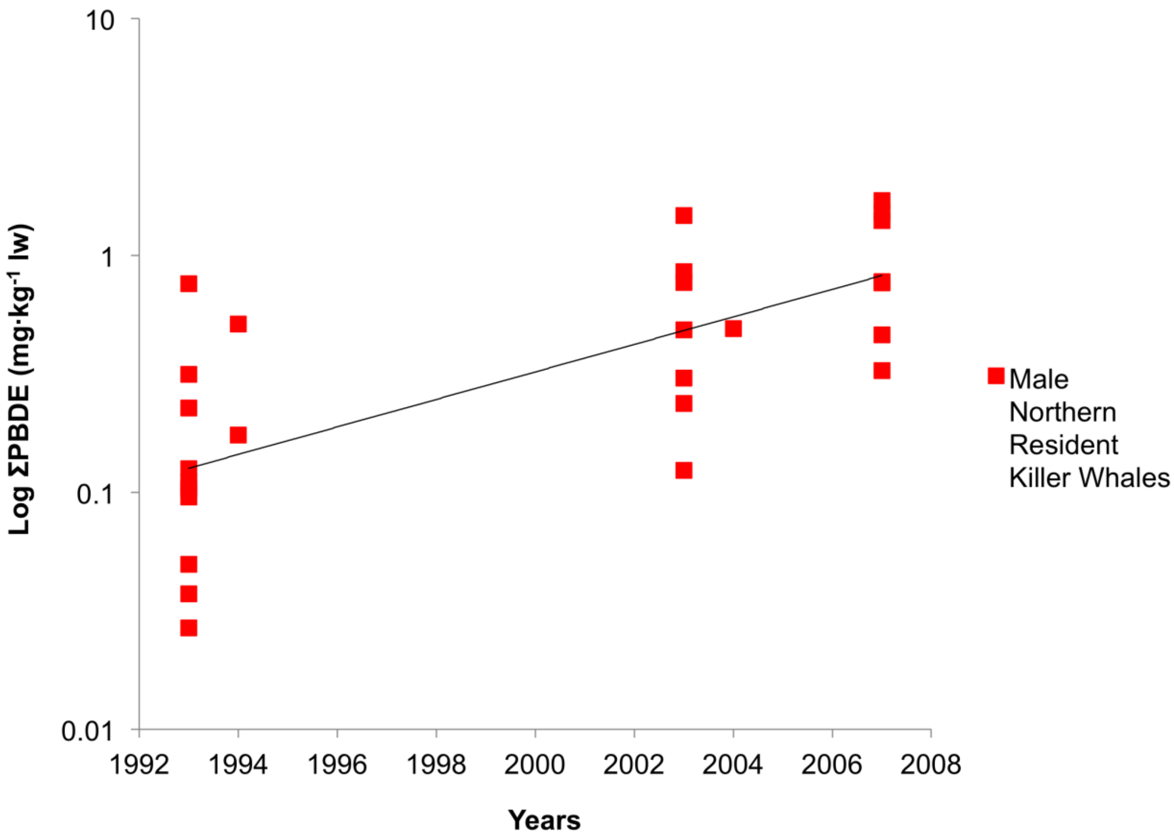

Figure 4.19 Log concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in male northern resident killer whales in units of $mg \cdot kg^{-1}$ lipid weight (lw; ■) versus time in years. The solid line represents the linear regression between the log-transformed concentrations of ΣPBDEs in killer whales and time.

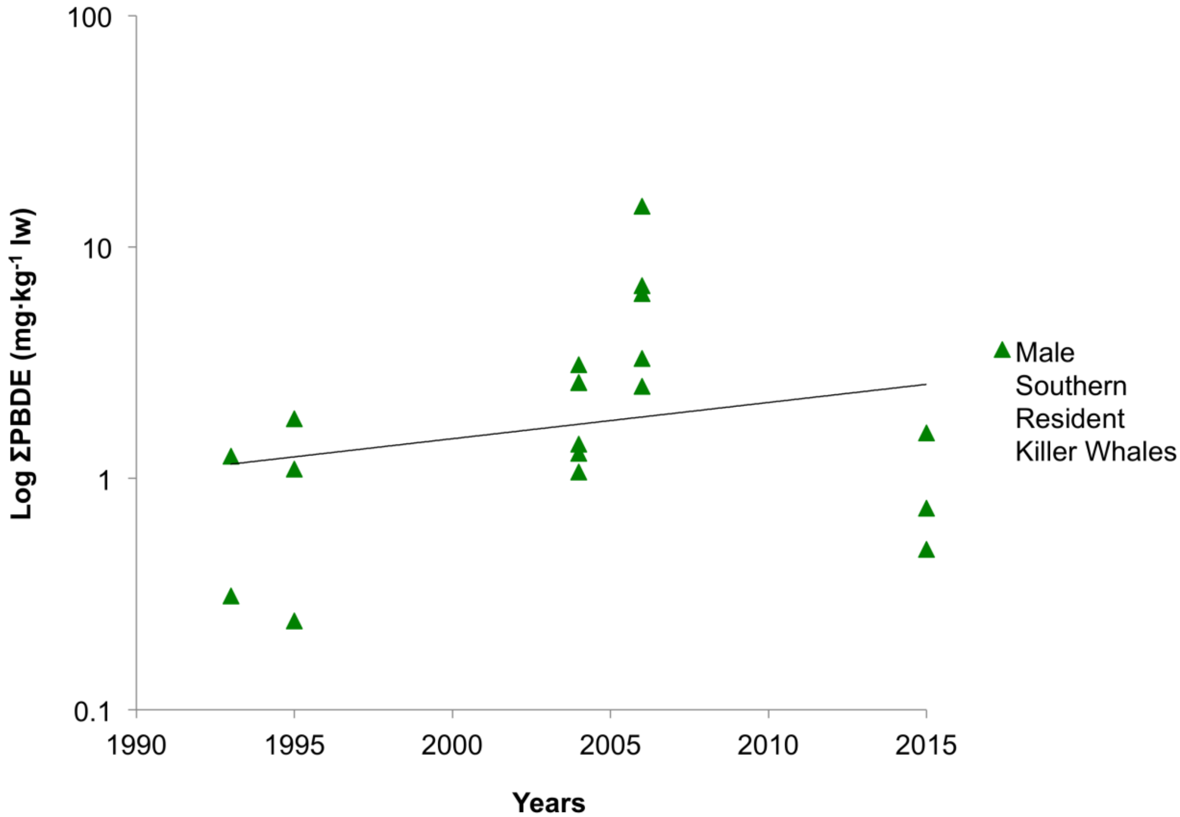

Figure 4.20 **Log concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in male southern resident killer whales in units of mg·kg^{-1} lipid weight (lw; ▲) versus time in years. The solid line represents the linear regression between the log-transformed concentrations of ΣPBDEs in killer whales and time.**

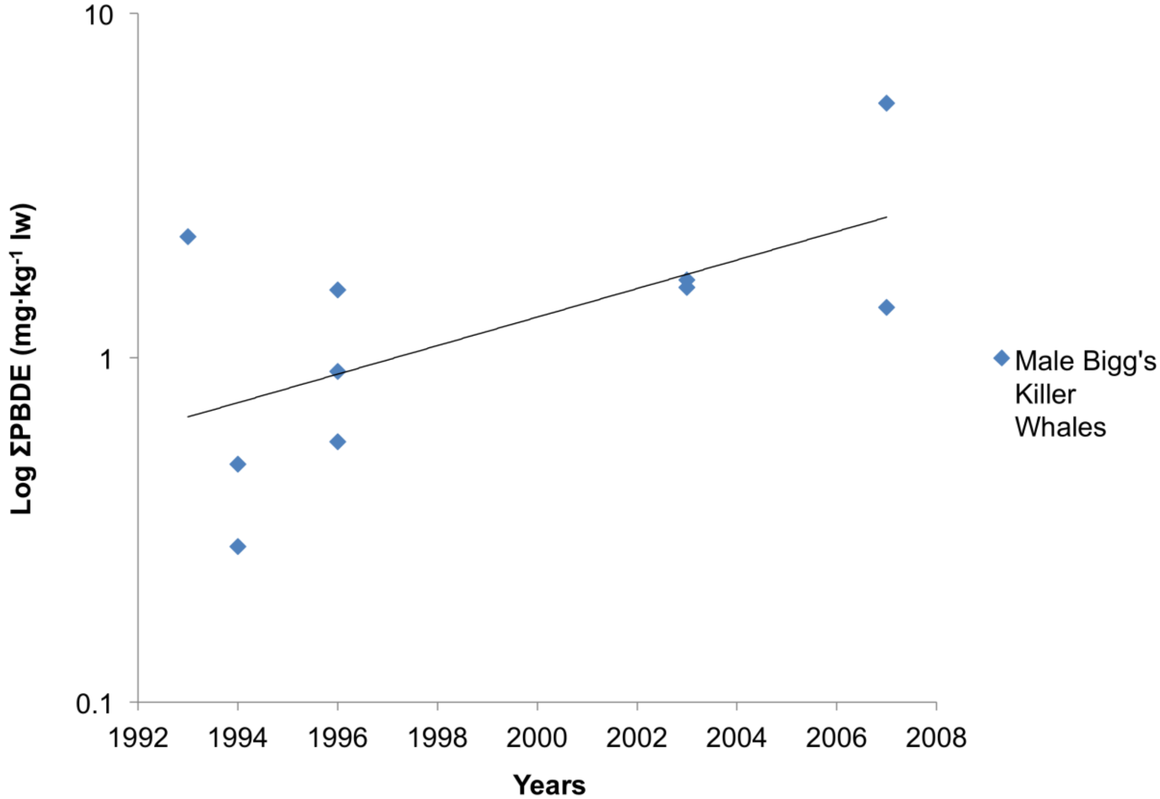

Figure 4.21 **Log concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in male Bigg's killer whales in units of mg·kg^{-1} lipid weight (lw; ◆) versus time in years. The solid line represents the linear regression of the log transformed concentrations of ΣPBDEs in killer whales and time.**

Table 4.9 **Outcomes of the linear regression of the logarithm of the concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in male killer whales over time. The analyses are presented for each ecotype – northern resident (NR), southern resident (SR) and Bigg's.**

Sex	Ecotype	Linear Regression	r^2	P-value
Male	NR	Log(ΣPBDE) = -117 + 0.0581*Year	0.508	<0.000100
Male	SR	Log(ΣPBDE) = -26.8 + 0.0135*Year	0.0471	0.372
Male	Bigg's	Log(ΣPBDE) = -82.7 + 0.0414*Year	0.382	0.0570

Running a similar linear regression for female killer whales showed that there was a significant increase in the log concentration of ΣPBDEs over time in both female northern resident killer whales (Figure 4.22; SE = 0.229, p = 0.0137) and female Bigg's killer whales

(Figure 4.24; SE = 0.0260, p = 0.0110). An insufficient number of female southern resident killer whales was measured for ΣPBDEs over time (only n = 1 in 2006 and then n = 6 in 2015), and therefore a linear regression analysis was not conducted (Figure 4.23). Results of the linear regression analyses of the concentrations of ΣPBDEs in female killer whales over time are depicted in Table 4.10.

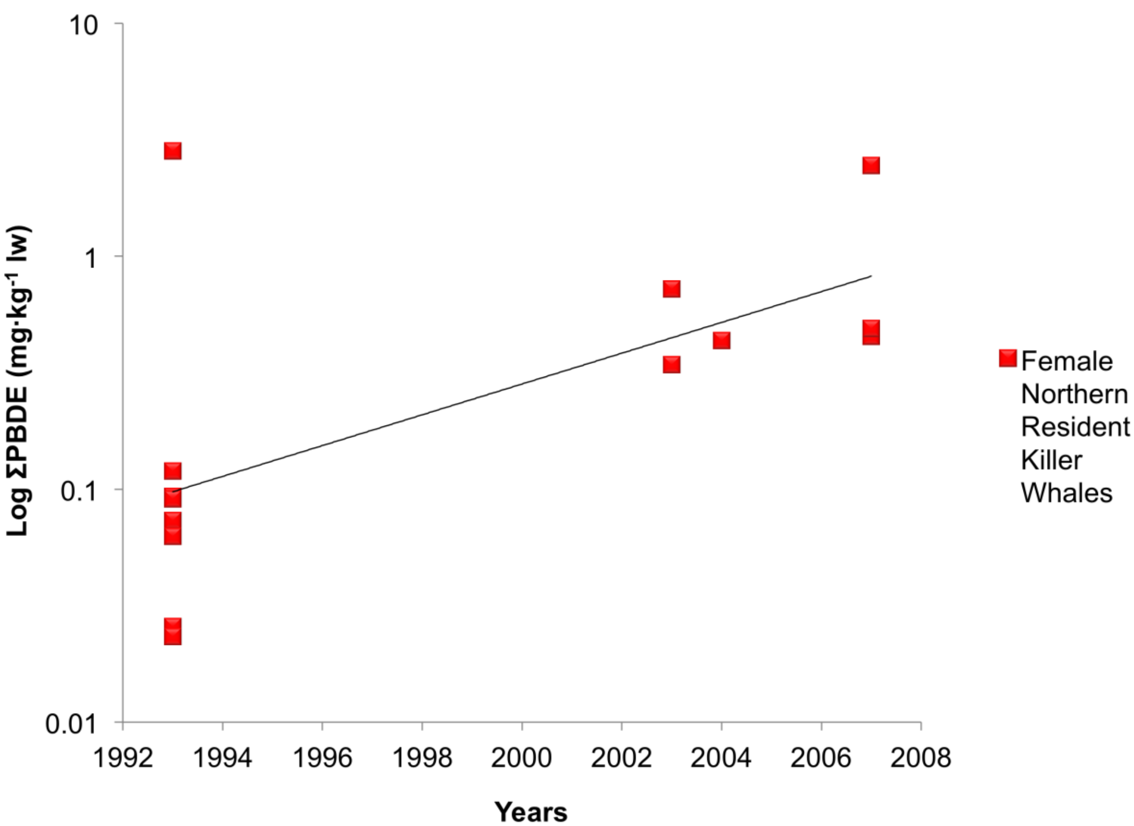

Figure 4.22 **Log concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in female northern resident killer whales in units of mg·kg^{-1} lipid weight (lw; ■) versus time in years. The solid line represents the linear regression between the log-transformed concentrations of ΣPBDEs in killer whales and time.**

Figure 4.23 **Log concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in female southern resident killer whales in units of mg·kg^{-1} lipid weight (lw; ■) versus time in years.**

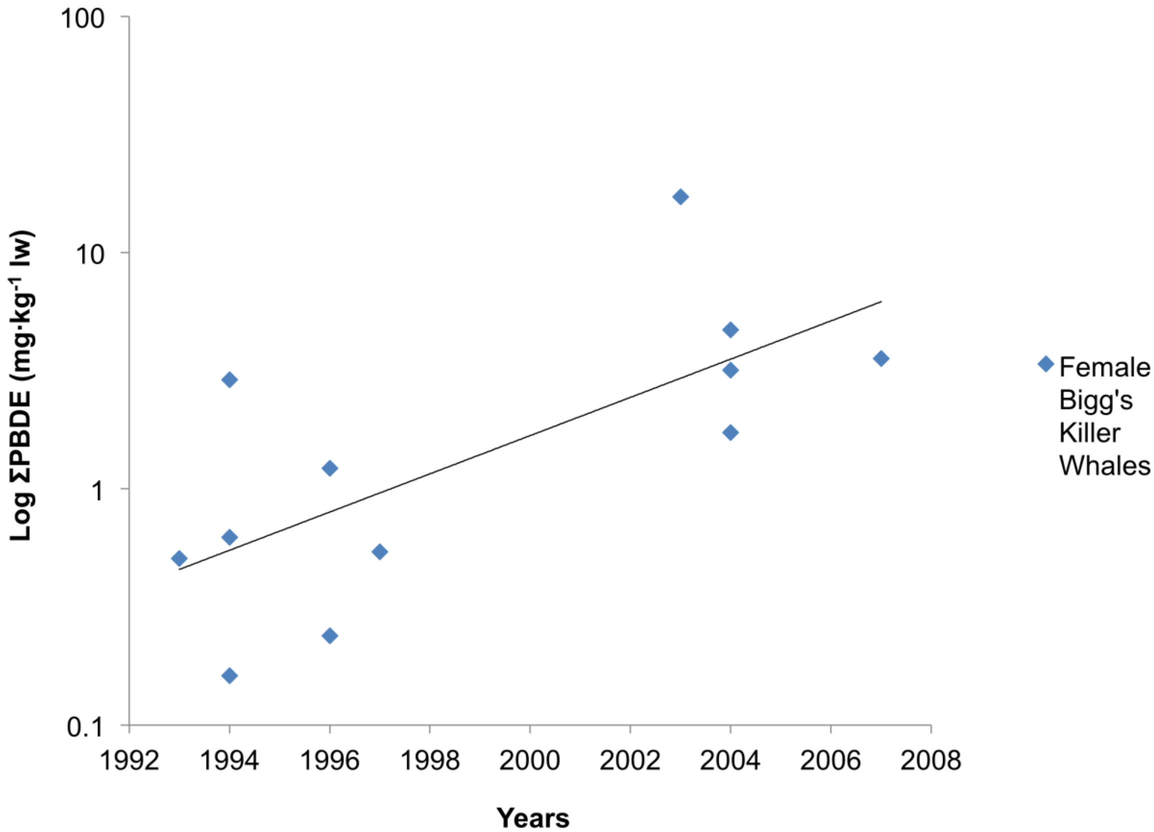

Figure 4.24 **Log concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in female Bigg's killer whales in units of mg·kg^{-1} lipid weight (lw; ◆) versus time in years. The solid line represents the linear regression between the log-transformed concentrations of ΣPBDEs in killer whales and time.**

Table 4.10 **Outcomes of the linear regression of the logarithm of the concentrations of total polybrominated diphenyl ethers (ΣPBDEs) in female killer whales over time. The analyses are presented for each ecotype – northern resident (NR), southern resident (SR) and Bigg's.**

Sex	Ecotype	Linear Regression	r^2	P-value
Female	NR	Log(ΣPBDE) = -133 + 0.066*Year	0.410	0.0137
Female	SR	N/A	N/A	N/A
Female	Bigg's	Log(ΣPBDE) = -162 + 0.0810*Year	0.493	0.0110

There are possible confounding factors that may have affected the linear regression analysis of ΣPBDEs in killer whales, such as the life history exposure, weight, calving order, sex and age of individual killer whales.

The linear regression analyses for both male and female killer whales showed that there were significant changes in the concentrations of ΣPBDEs in killer whales from 1993 to 2015. A possible reason for this increase in concentrations of ΣPBDEs over time is that inputs of PBDEs into the habitat of killer whales are increasing. To investigate if ΣPBDEs had any elimination rates that could be affecting these changes in concentrations in killer whales over time, the half-life of ΣPBDEs in killer whales was calculated.

The half-lives of ΣPBDEs in killer whales are depicted in Table 4.11. The half-life calculations used were based upon those of Alava et al. [64]. One factor included in the half-life calculations was k_m, the metabolic transformation rate. Harju et al. [69]'s metabolic degradation half-lives were initially considered for the present study's half-life calculations, as their degradation rates are the only known available in-vitro measurements for PBDE congeners. Harju et al. [69] found that metabolic degradation rates for PBDE congeners in Wistar-Unilever ranged from 4 mins to 170 mins. These short metabolic degradation rates were concluded by the present study to not be realistic in the natural environment of a killer whale, however, and thus Harju et al. [69]'s metabolic rates were not used for the calculations of PBDE half-life in the present study, and k_m was instead set to zero.

In adult male killer whales, the half-life of PBDEs was determined to be 11.9 years whereas in female adult killer whales, PBDEs were found to have a half-life of 2.06 years. PBDEs in juvenile killer whales were found to have a half-life of 2.88 years and in killer whale calves, PBDEs had a half-life of 0.482 years.

Table 4.11 Half-lives (in years) of total polybrominated diphenyl ethers (ΣPBDEs) in killer whale blubber, calculated for different life stages of killer whales: adult male, adult female, juvenile and calf.

Killer Whale Life Stage	Half-life (years)
Adult Male	1.19E+01
Adult Reproductive Female	2.06E+00
Juvenile	2.88E+00
Calf	4.82E-01

The half-lives of PBDEs in killer whales were similar in length to the half-lives of PCBs in killer whales; however, where concentrations of PCBs were not found to change over time, PBDEs were found to change significantly over time in killer whales. The time period explored for concentrations of PBDEs ranged from 13 years to 22 years. According to the half-life calculations, a decrease in PBDEs should have been observed if there were no inputs of PBDEs into the killer whale's environment. Instead, however, an increase in concentrations of PBDEs was found in female Bigg's, and female and male northern resident killer whales. It is therefore possible that there has been an increase in the concentrations of PBDEs entering the environment of the Salish Sea killer whales.

4.2.4. Age – PBDEs – Killer Whales

Linear Regression

To investigate any confounding effects, age was plotted against year of sampling for both female (Figures 4.25 – 4.27) and male killer whales (Figures 4.28 – 4.30).

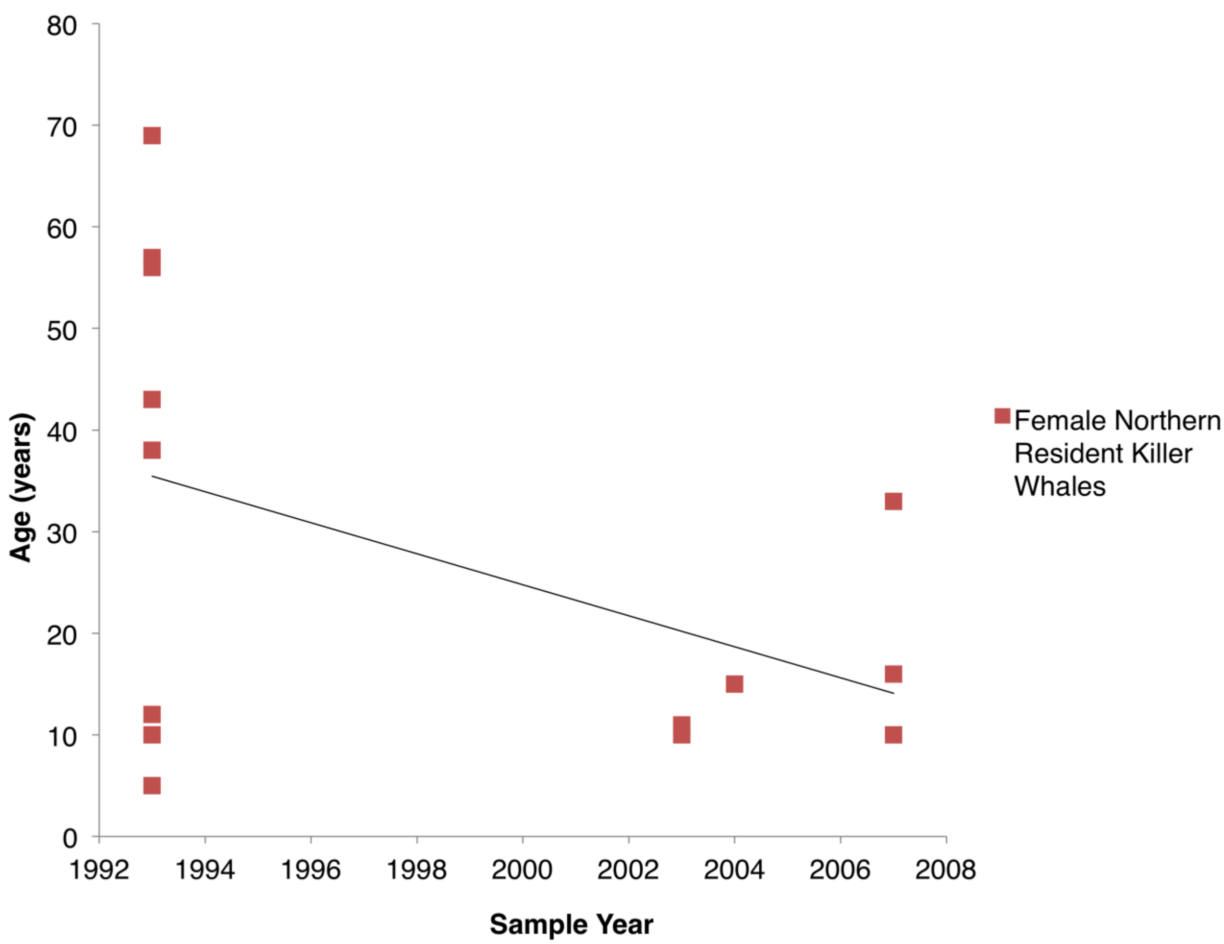

Figure 4.25 The age of female northern resident killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which

each killer whale was sampled. The solid line gives the linear regression of killer whale age versus sample year.

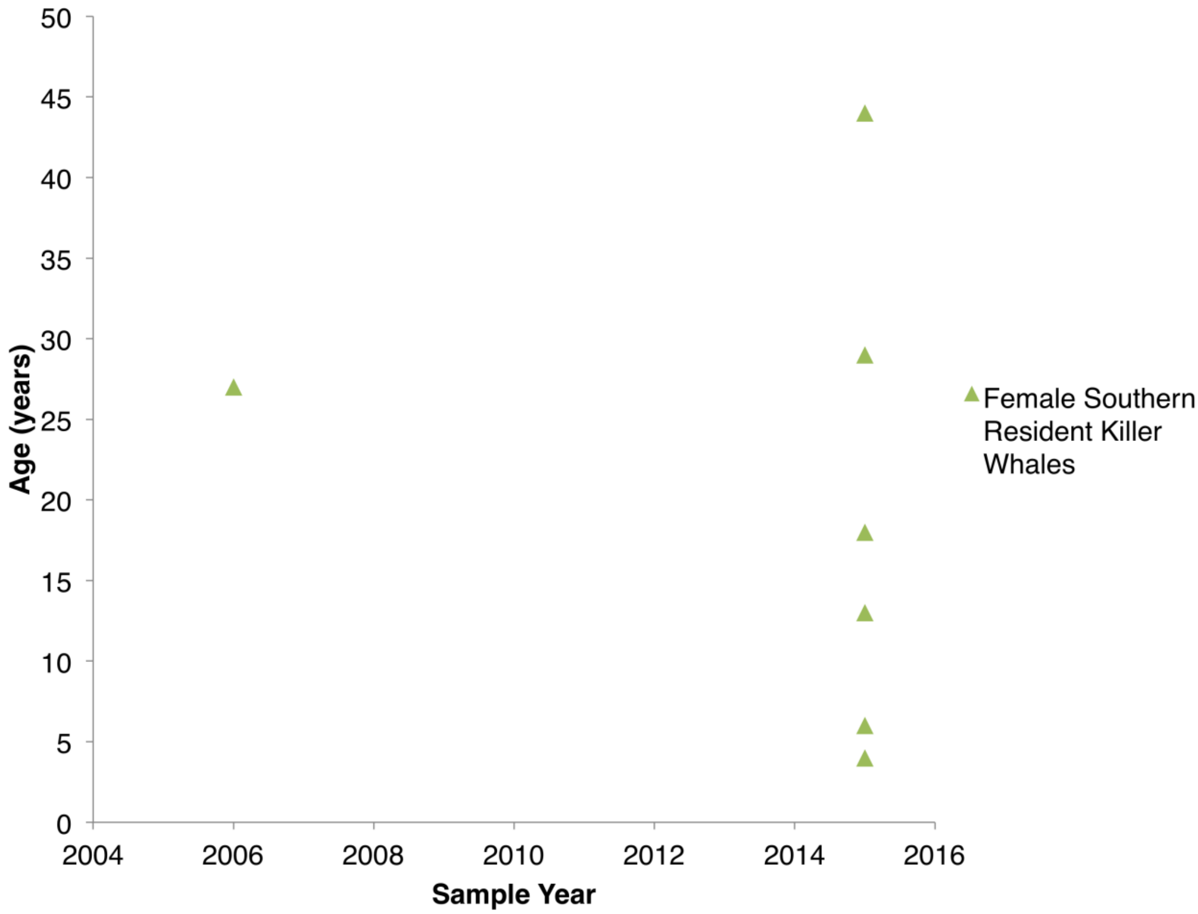

Figure 4.26 **The age of female southern resident killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled.**

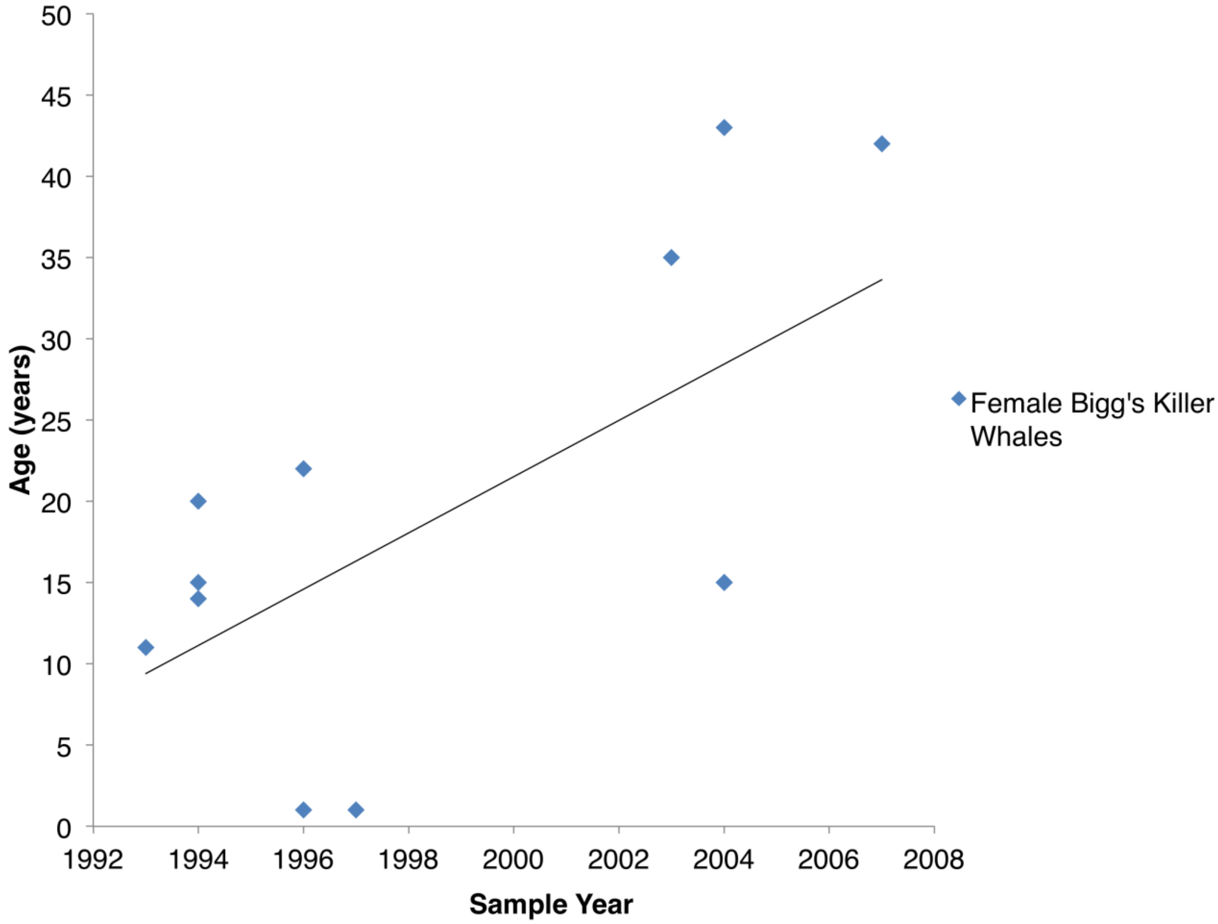

Figure 4.27 **The age of female Bigg's killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The solid line gives the linear regression of killer whale age versus sample year.**

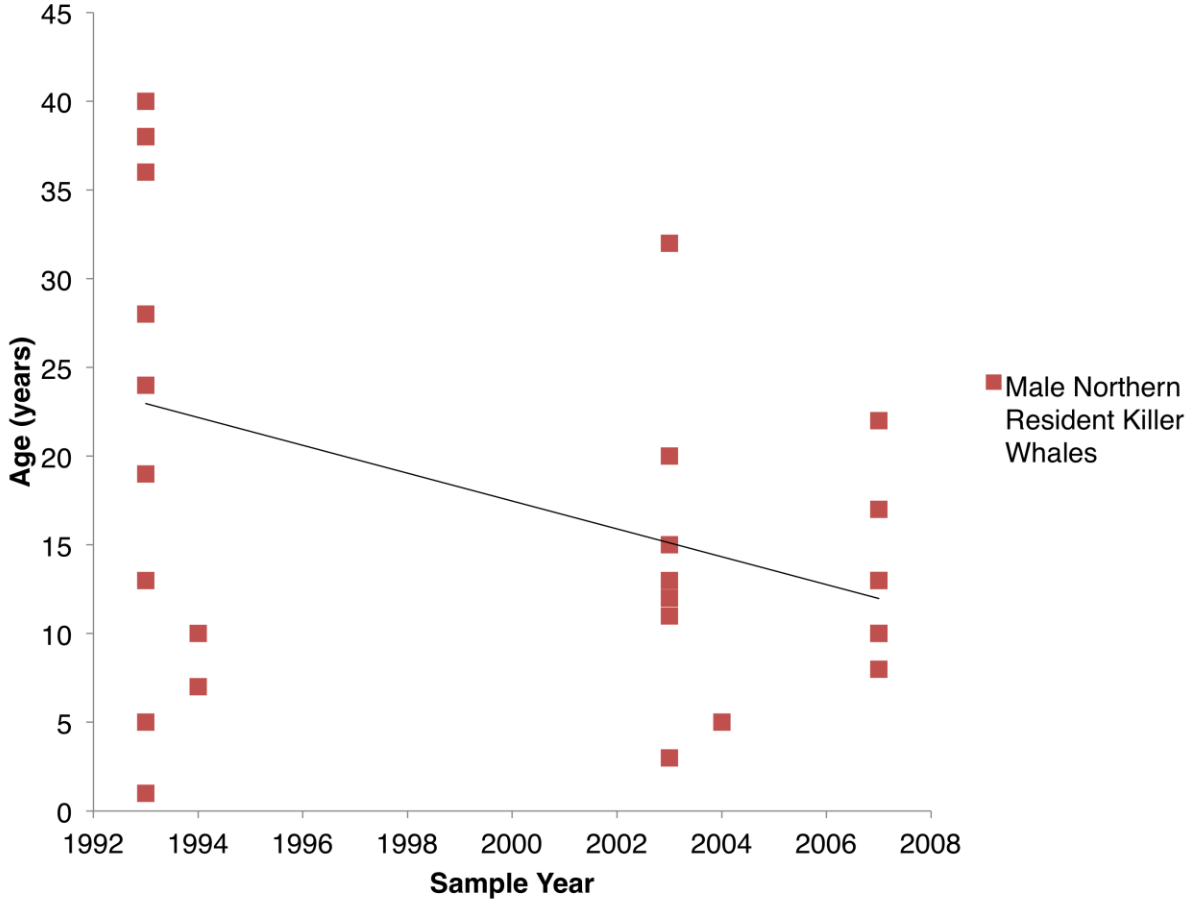

Figure 4.28 **The age of male northern resident killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The solid line gives the linear regression of killer whale age versus sample year.**

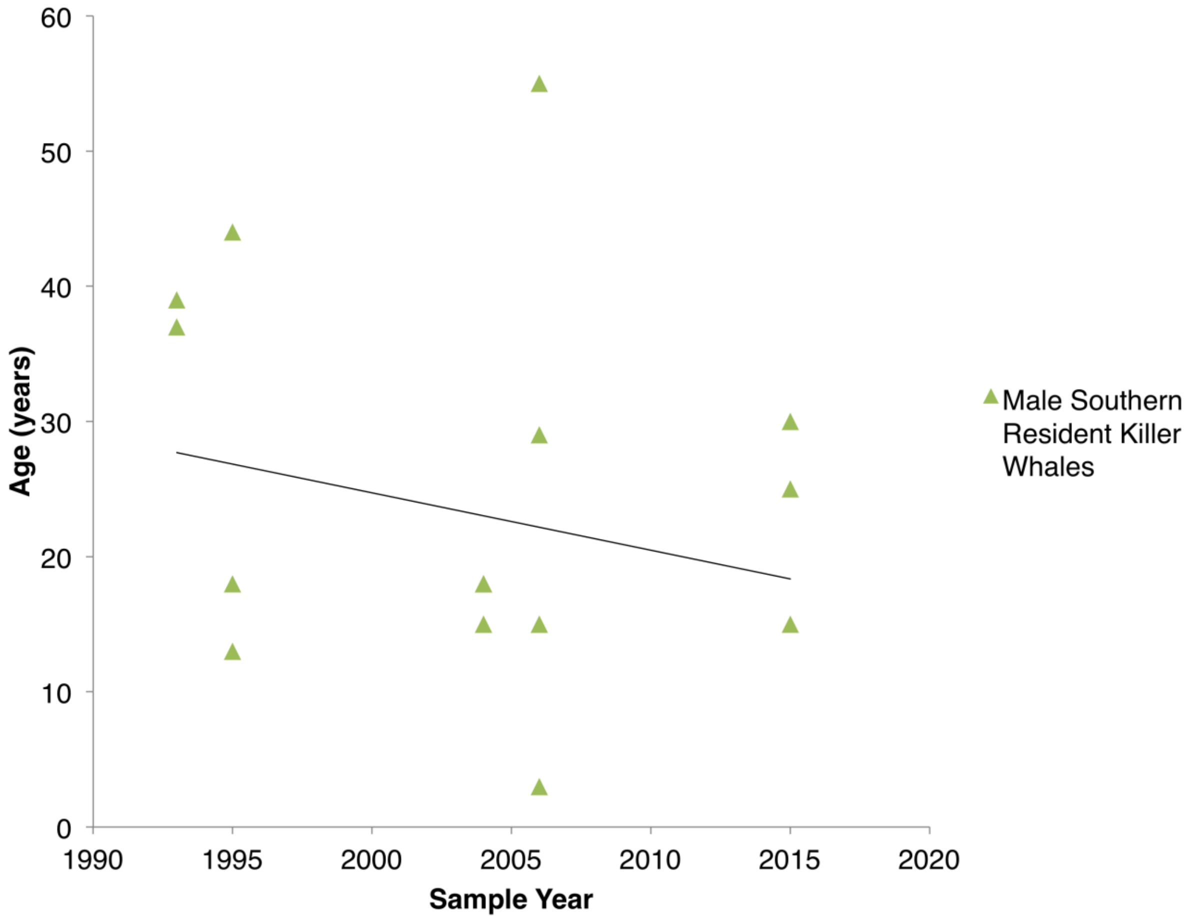

Figure 4.29 **The age of male southern resident killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The solid line gives the linear regression of killer whale age versus sample year.**

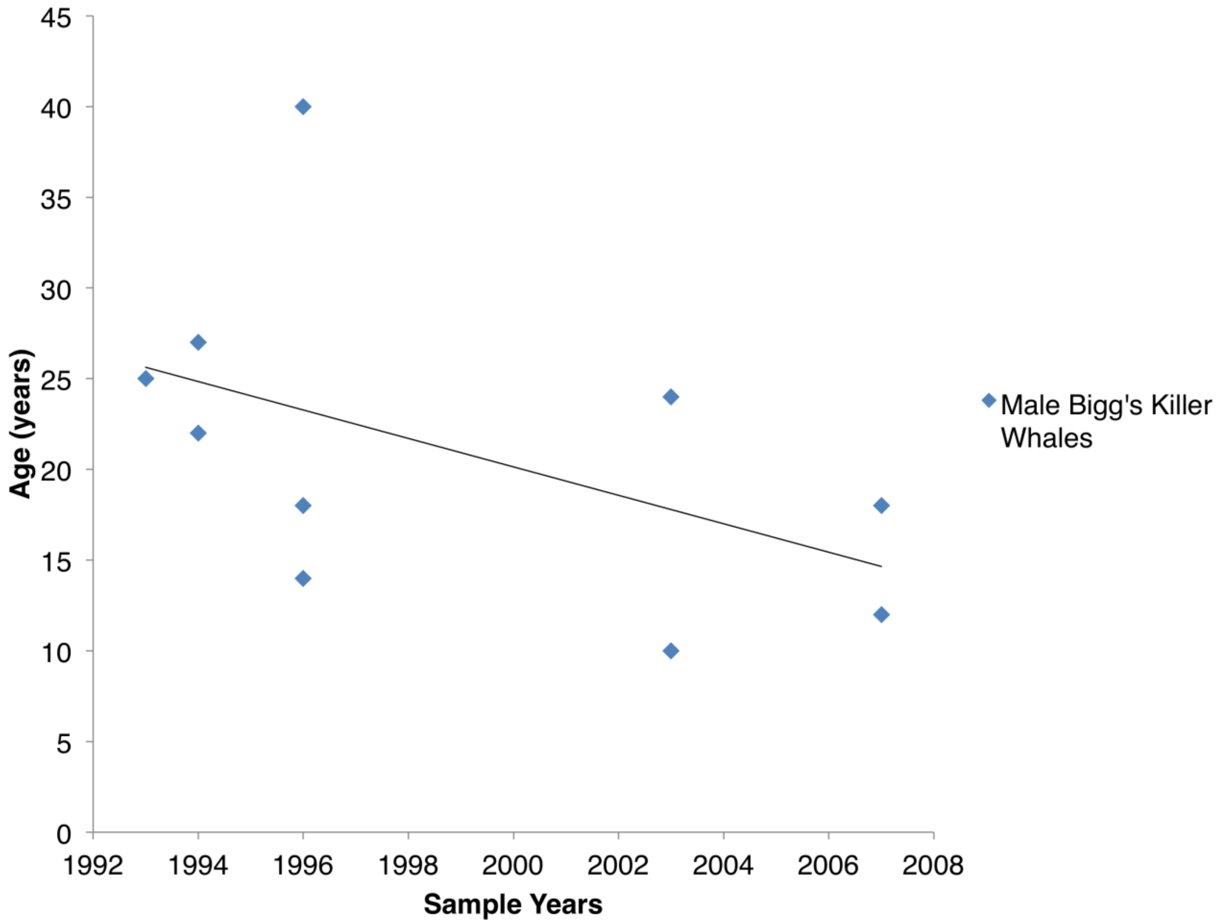

Figure 4.30 The age of male Bigg's killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The solid line gives the linear regression of killer whale age versus sample year.

Outcomes of the linear regression of age versus year in which PBDEs were sampled in male and female killer whales are given in Table 4.12. There were no significant relationships found between age and sampling year for female and male northern resident killer whales, or for male southern resident killer whales. No linear regression analysis was conducted for female southern resident killer whales sampled for ΣPBDEs, due to an insufficient sample size (only n = 1 in 2006, and then n = 6 in 2015). There was, however, a significant positive relationship between age and sample year for female Bigg's killer whales (SE = 6.70E-01, p = 0.0272), and a significant negative relationship between age and sample year for male Bigg's killer whales (SE = 0.490, p = 0.148).

Table 4.12 Results of linear regressions comparing the age of Salish Sea killer whales and the year in which each whale was sampled for polybrominated diphenyl ethers (PBDEs). Analyses were conducted separately by sex and by ecotype – northern resident (NR), southern resident (SR) and Bigg's.

Sex	Ecotype	Linear Regression	r^2	P-value
Female	NR	Age = 3078 - 1.53*Year	2.05E-01	0.104
Female	SR	-	-	-
Female	Bigg's	Age = -3441 + 1.73*Year	4.00E-01	0.0272
Male	NR	Age = 1588 - 0.785*Year	5.39E-01	0.158
Male	SR	Age = 875 - 0.425*Year	5.30E-02	0.343
Male	Bigg's	Age = 1589 - 0.784*Year	9.78E-01	0.00130

Mean age was also plotted against year in which PBDEs were sampled for both female (Figures 4.31 – 4.32) and male killer whales (Figures 4.33 – 4.35).

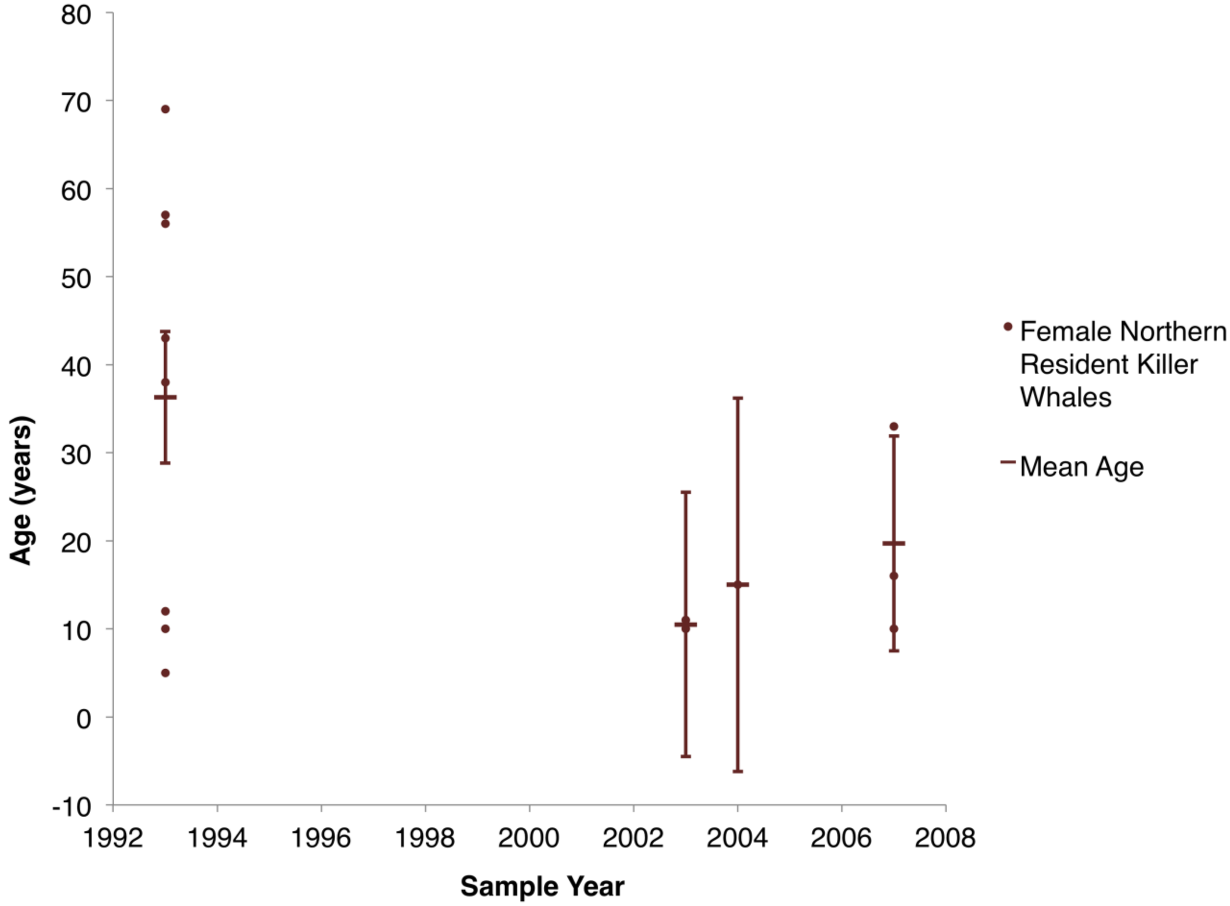

Figure 4.31 **The age of female northern resident killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of killer whales in that sample year.**

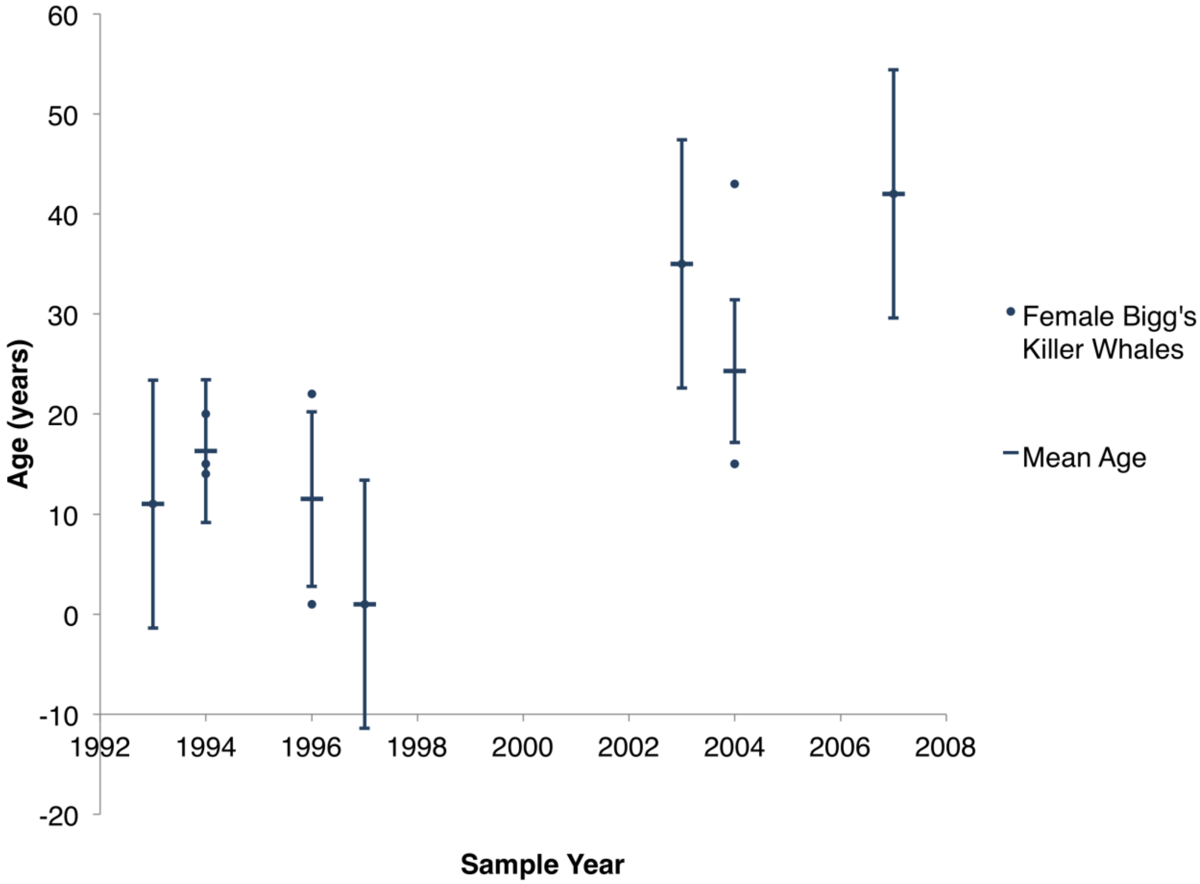

Figure 4.32 **The age of female Bigg's killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of killer whales in that sample year.**

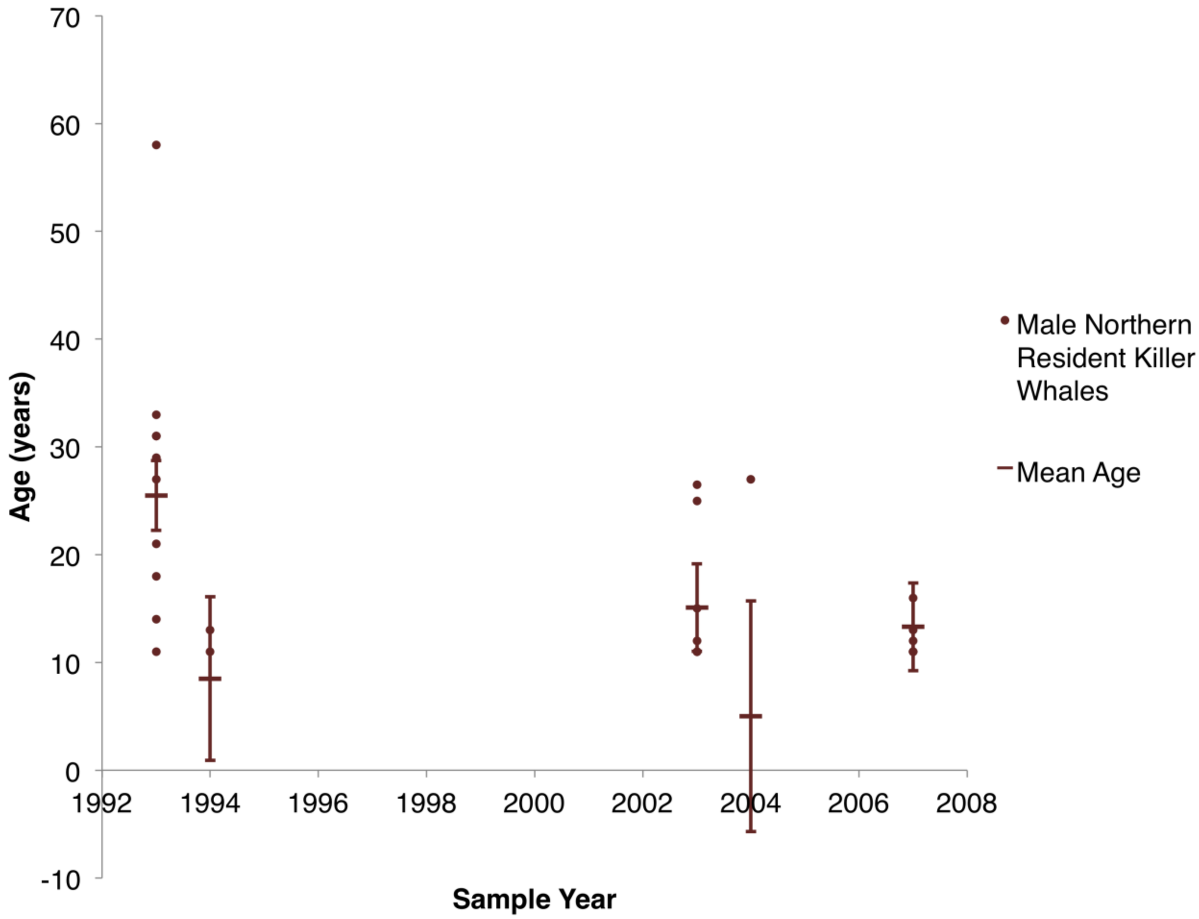

Figure 4.33 The age of male northern resident killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of killer whales in that sample year.

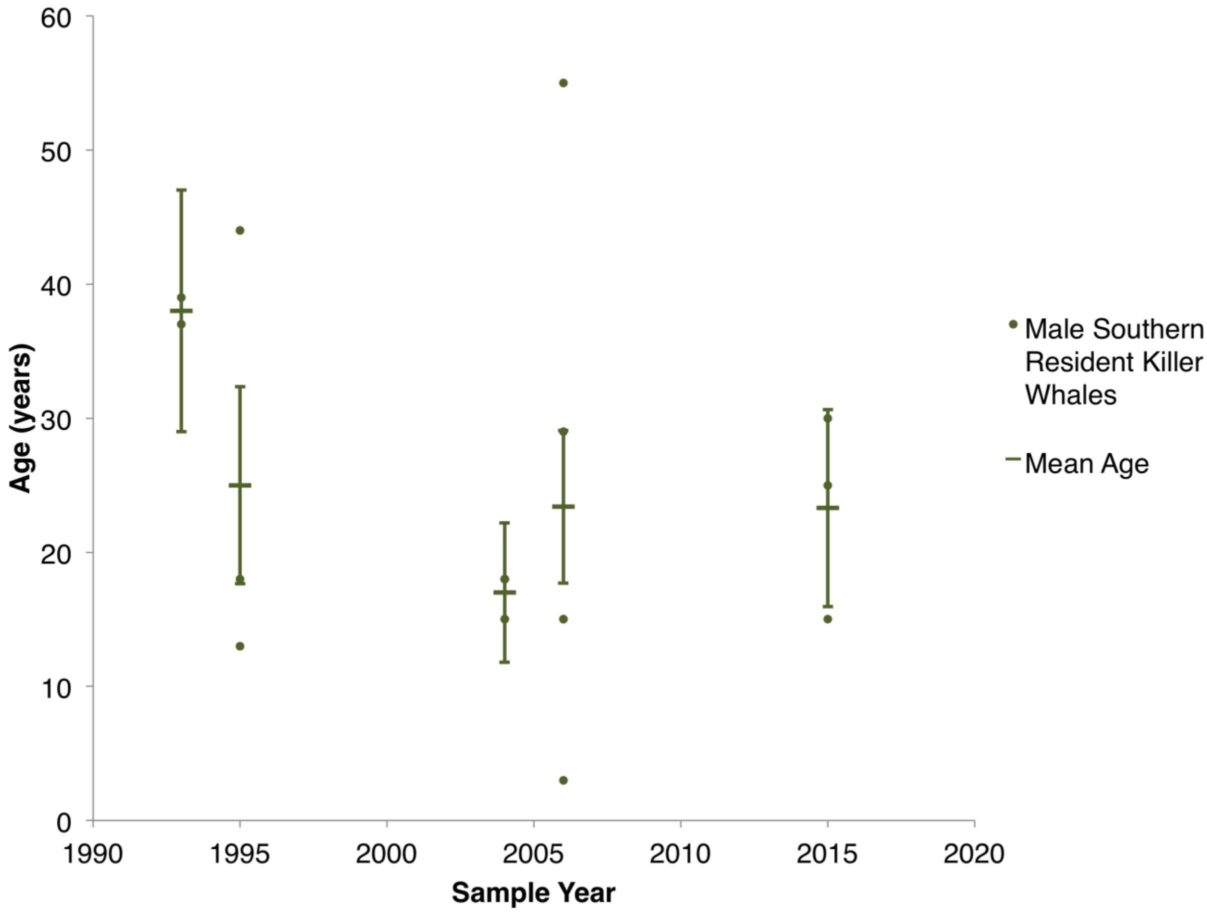

Figure 4.34 **The age of male southern resident killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of killer whales in that sample year.**

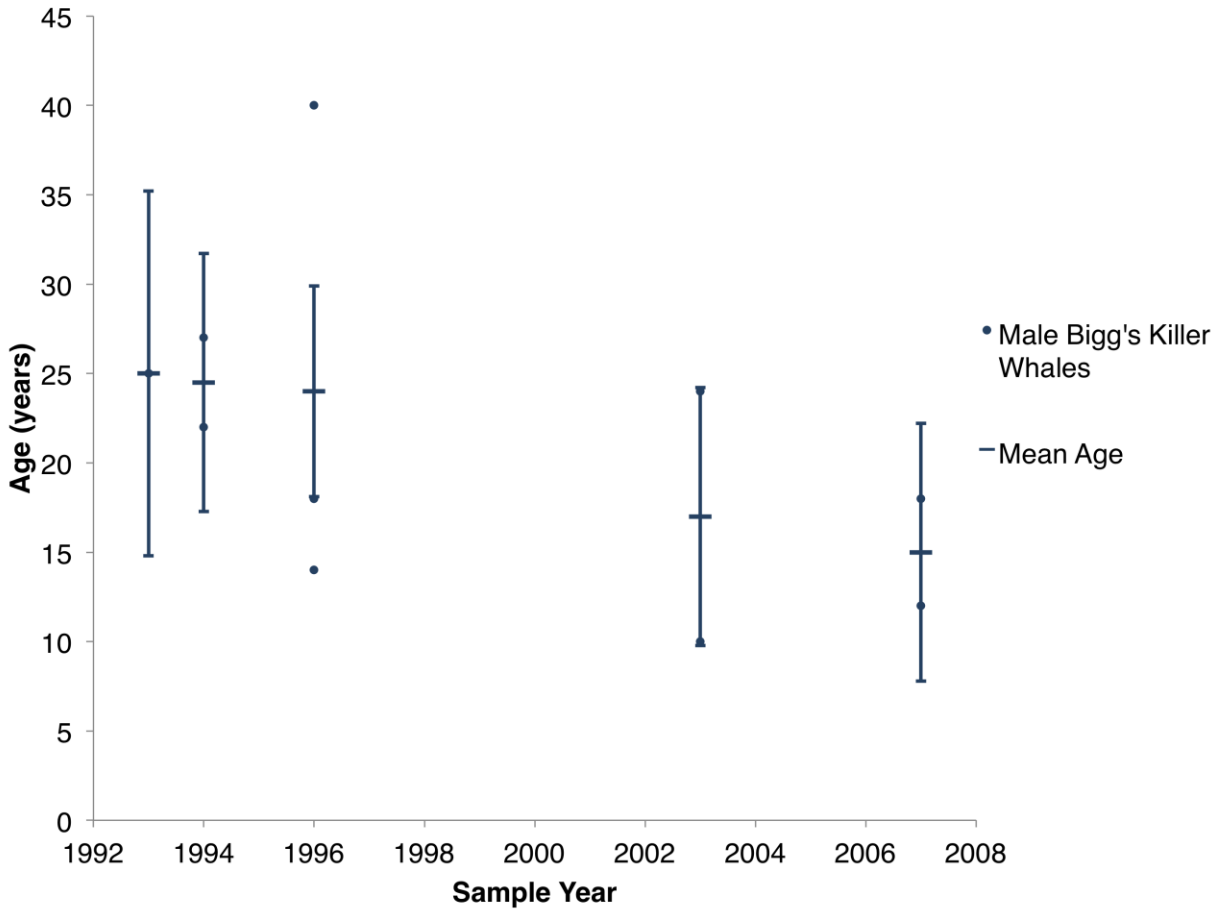

Figure 4.35 The age of male Bigg's killer whales sampled for total polybrominated diphenyl ethers (ΣPBDEs) plotted against the year in which each killer whale was sampled. The horizontal lines represent the mean age of killer whales in that sample year.

An ANOVA showed that the mean ages of both females and males did not significantly differ among the years in which PBDEs were sampled (Table 4.13). No ANOVA results were obtained for female southern resident killer whales due to insufficient data (see Appendix G for mean ages for each sampled year).

Table 4.13 Results of ANOVA comparing the mean age of Salish Sea killer whales in each year during which ΣPBDEs were sampled. Tests were conducted separately by sex and by ecotype – northern resident (NR), Bigg's and southern resident (SR).

Sex	Ecotype	F Ratio	P-value
Female	NR	1.14E+00	0.38
Female	Bigg's	1.52E+00	0.332
Male	NR	2.55E+00	0.0667
Male	SR	1.05E+00	0.419
Male	Bigg's	4.12E-01	0.795

For female Bigg's killer whales, both age and concentrations of PBDEs were found to significantly increase over time. It can be presumed that older animals have had greater exposure to concentrations of PBDEs than have younger animals; this is to be expected, due to the longer life history of older whales. Ross et al.[1] explained that non-reproductive females are also known to exhibit increases in contaminant concentrations over time. The significant increase in age of female Bigg's killer whales over time, however, shows that age could be a confounding factor in the relationship between concentrations of PBDEs and time in these whales. For female Bigg's killer whales, concentrations of PBDEs may not truly be increasing over time.

The age of male Bigg's killer whales also significantly changed over time; however, concentrations of PBDEs were not found to significantly change over time in male Bigg's killer whales. Additionally, age did not significantly change for male and female northern resident whales, or for southern resident killer whales. The mean age of killer whales for all sample periods also did not change significantly among the sample periods. For these killer whale ecotypes and sexes (all male ecotypes, and female northern and southern resident killer whales), age was found to not be a confounding factor for concentrations of PBDEs changing over time in killer whales. Rayne et al.[8] also found no relationship between age and PBDE concentrations in killer whales, and instead concluded that PBDE may be a new contaminant of concern for the health of killer whales. The present study further strengthens the argument that PBDEs are a contaminant of concern for the health of killer whales, especially northern and southern resident killer whales. Indeed, PBDEs must be monitored due to their increasing concentrations in killer whales over time.

4.2.5. Other Emerging Pollutants in Killer Whales

The blubber tissues sampled in 2015 were tested for various chlorinated and brominated flame-retardants. This was the first time these chemicals were measured in the Salish Sea killer whales. Of the group of chemicals that were measured (Appendix A), several emerging pollutants were detected, including ΣHBCDD, Dechlorane, Dechlorane Plus, Dec 602, PBT, 1,2-DiBB, and TBCT. Due to this being the first time these chemicals have been measured, a temporal comparison could not be done.

Concentration measurements of ΣHBCDD in killer whales included the sum of the three isomers of HBCDD (alpha, beta, and gamma). The geometric means for all killer whales sampled in 2015 are presented in Table 4.14, as are the geometric mean for females and male killer whales.

Table 4.14 Geometric means and their corresponding upper and lower standard deviations (SD) for total hexabromocyclododecane isomers (ΣHBCDD) in killer whales sampled in 2015.

Contaminant	ΣHBCDD
Geometric Mean ($mg{\cdot}kg^{-1}$ lw)	6.79E-02
Upper SD ($mg{\cdot}kg^{-1}$ lw)	9.67E-02
Lower SD ($mg{\cdot}kg^{-1}$ lw)	4.77E-02
Female Geometric Mean ($mg{\cdot}kg^{-1}$ lw)	5.82E-02
Female Upper SD ($mg{\cdot}kg^{-1}$ lw)	7.08E-02
Female Lower SD ($mg{\cdot}kg^{-1}$ lw)	4.79E-02
Male Geometric Mean ($mg{\cdot}kg^{-1}$ lw)	9.24E-02
Male Upper SD ($mg{\cdot}kg^{-1}$ lw)	1.43E-01
Male Lower SD ($mg{\cdot}kg^{-1}$ lw)	5.96E-02

One previous study explored the concentrations of ΣHBCDD in harbour porpoises located in different European seas, and found that ΣHBCDD in harbour porpoises along the Irish coast were present at a concentration of 2.9 $mg{\cdot}kg^{-1}$ lw [70]. This value is higher than the values found for killer whales in the Salish Sea, showing that concentrations of HBCDD are lower in killer whales than in harbour porpoises off the Irish coast. This could possibility mean that the

Salish Sea is not experiencing as many inputs of HBCDD into its waters, and therefore HBCDD is not likely to be a main contaminant of concern for the health of the Salish Sea killer whales.

The geometric means and associated standard deviations for the concentrations of Dechlorane, Dechlorane plus, Dec 602, 1,2-DiBB, PBT, and TBCT measured in the blubber of Salish Sea killer whales sampled in 2015 are shown in Table 4.15. Other emerging contaminants were tested but were not detected (see Appendix A for a complete list of all contaminants tested).

Table 4.15 **Geometric mean concentrations of Dechlorane, Dechlorane Plus, Dechlorane 602 (Dec 602), 1,2-dibromobenzene (1,2-DiBB), pentabromotoluene (PBT) and tetrabromo-o-chlorotoluene (TBCT) in all killer whales sampled in 2015. Means are also given separately for male and female killer whales sampled in 2015. Units are stated in mg·kg^{-1} lipid weight (lw). Corresponding upper and lower standard deviations (SD) are given.**

Contaminant	Geometric Mean (mg·kg^{-1} lw)	Upper SD	Lower SD	Female Geometric Mean (mg·kg^{-1} lw)	Female Upper SD	Female Lower SD	Male Geometric Mean (mg·kg^{-1} lw)	Male Upper SD	Male Lower SD
Dechlorane (Mirex)	6.69E-02	1.18E-01	3.79E-02	6.21E-02	1.04E-01	3.70E-02	7.76E-02	1.65E-01	3.65E-02
Dechlorane Plus	6.58E-03	1.39E-02	3.12E-03	5.53E-03	1.33E-02	2.30E-03	9.32E-03	1.09E-02	7.97E-03
Dec 602	3.06E-03	4.94E-03	1.90E-03	3.39E-03	4.96E-03	2.31E-03	2.51E-03	4.94E-03	1.28E-03
1,2-DiBB	6.06E-02	1.73E-01	2.12E-02	4.82E-02	1.69E-01	1.37E-02	9.55E-02	9.88E-02	9.24E-02
PBT	8.16E-03	2.20E-02	3.02E-03	6.74E-03	2.18E-02	2.08E-03	1.20E-02	1.80E-02	7.98E-03
TBCT	6.93E-03	1.53E-02	3.14E-03	5.03E-03	1.07E-02	2.36E-03	1.31E-02	1.94E-02	8.91E-03

This was the first time Dechlorane, Dechlorane plus, Dec 602, 1,2-DiBB, PBT, and TBCT were measured in Salish Sea killer whales. Several studies have investigated these chemicals in other marine mammals. For example, de la Torre et al. [71] investigated concentrations of Dechlorane (mirex), Dec 602, and Dechlorane Plus in Franciscana dolphins (*Pontoporia blainvillei*), which are found off of the Southern coast of Brazil.

The present study determined that Dechlorane (mirex) had the highest geometric mean concentration of all the investigated emerging pollutants in killer whales. Similarly, de la Torre et al. [71] found that concentrations of Dechlorane were the highest of all the contaminants that were investigated. The Franciscana dolphins had a lower geometric mean concentration (3.35E-02 $mg\cdot kg^{-1}$ lw) of Dechlorane than did the Salish Sea killer whales in the current study (6.67E-02 $mg\cdot kg^{-1}$ lw), indicating that killer whales may be experiencing higher contaminant concentrations of chemicals from the Dechlorane family than are other marine mammal species. The work of de la Torre et al. [71] was one of the few studies found to be researching chemical concentrations from the Dechlorane family in aquatic mammals. This highlights the lack of information concerning these flame-retardants, and more investigation is needed to understand if these contaminants have detrimental health effects on Salish Sea killer whales.

The ages of the female and male killer whales sampled in 2015 were plotted against the concentrations of the detected emerging pollutants (Figures 4.36 – 4.42), and corresponding linear regressions were conducted to determine if there were any significant relationships between age and contaminant concentrations. If significant relationships were found, age would be considered a sampling artefact for that data set. The results and p-values for the above mentioned linear regression analyses are presented in Table 4.16. The second youngest killer whale (L116, Age = 6, Sex = Male) in the 2015 dataset exhibited the highest concentrations of ΣHBCDD, TBCT, 1,2-DiBB, and Dechlorane. No significant relationships were found between the concentrations of emerging pollutants and age in either female or male killer whales (see Table 4.16).

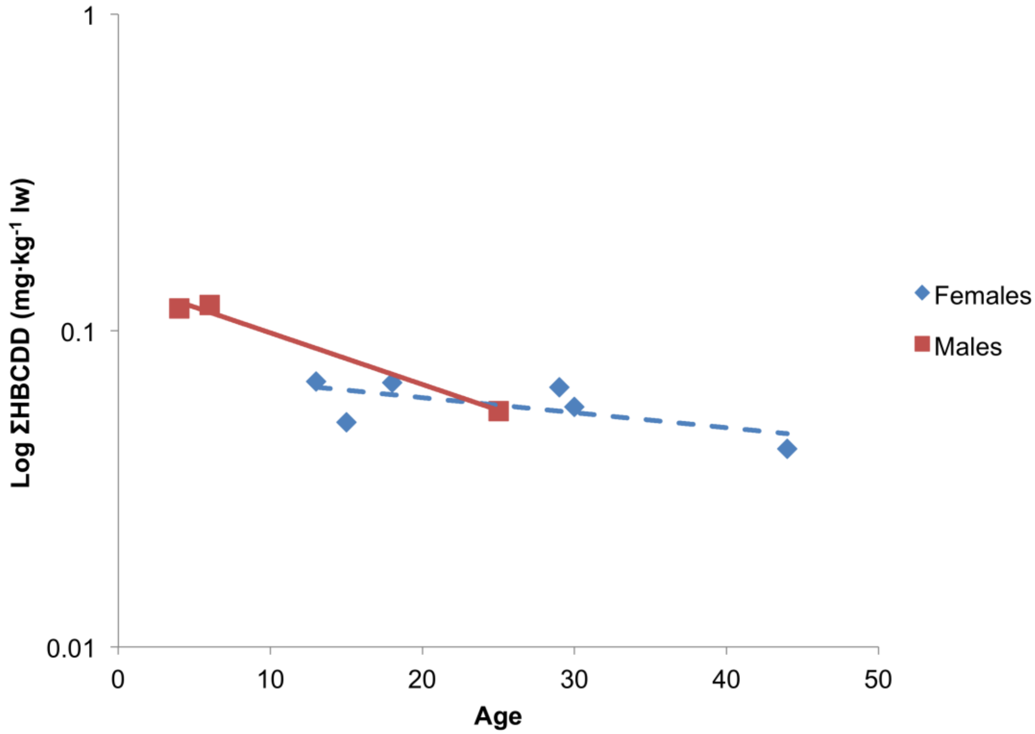

Figure 4.36 **The log-transformed concentrations of total hexabromocyclododecane isomers (ΣHBCDDs; mg·kg^{-1} lw) in southern resident killer whales as a function of the ages (in years) of killer whales sampled in 2015. Two linear regression analyses are shown, one for females (◆) and the other for males (■).**

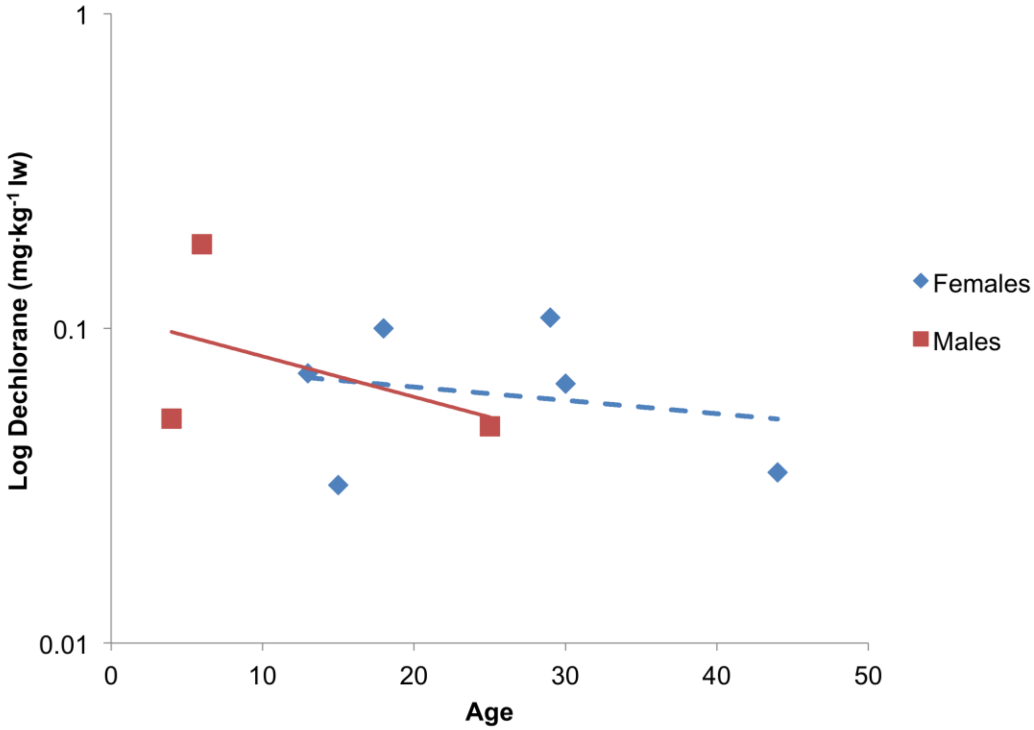

Figure 4.37 **The log-transformed concentrations of Dechlorane (mg·kg^{-1} lw) in southern resident killer whales as a function of the ages (in years) of killer whales sampled in 2015. Two linear regression analyses are shown, one for females (◆) and the other for males (■).**

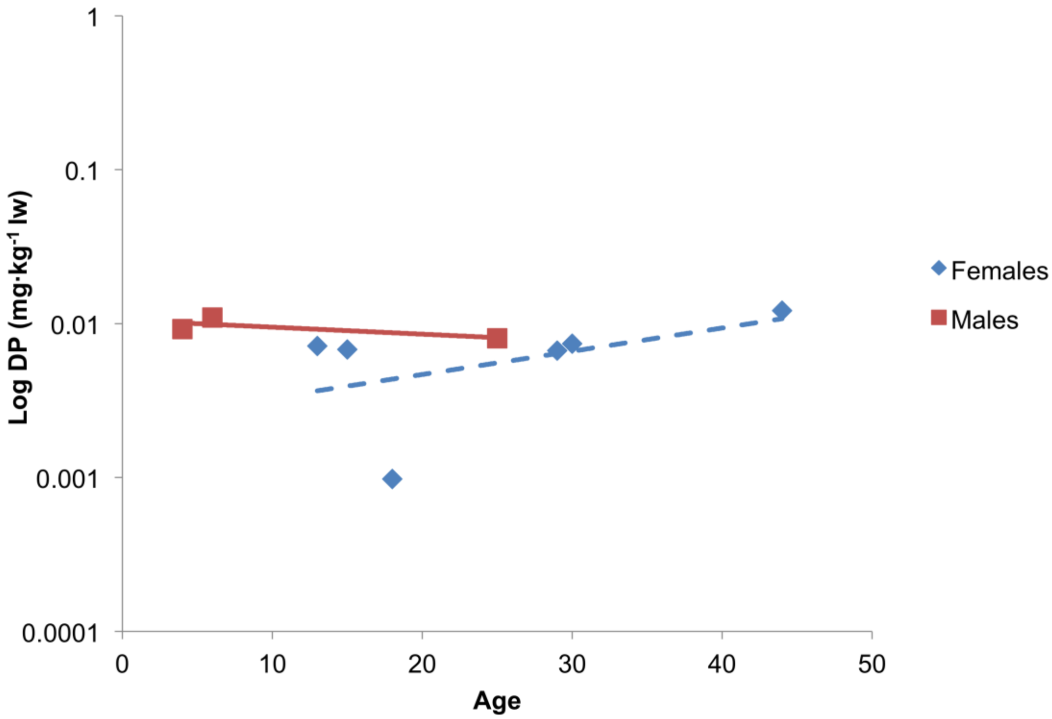

Figure 4.38 **The log-transformed concentrations of Dechlorane Plus (mg·kg^{-1} lw) in southern resident killer whales as a function of the ages (in years) of killer whales sampled in 2015. Two linear regression analyses are shown, one for females (◆) and the other for males (■).**

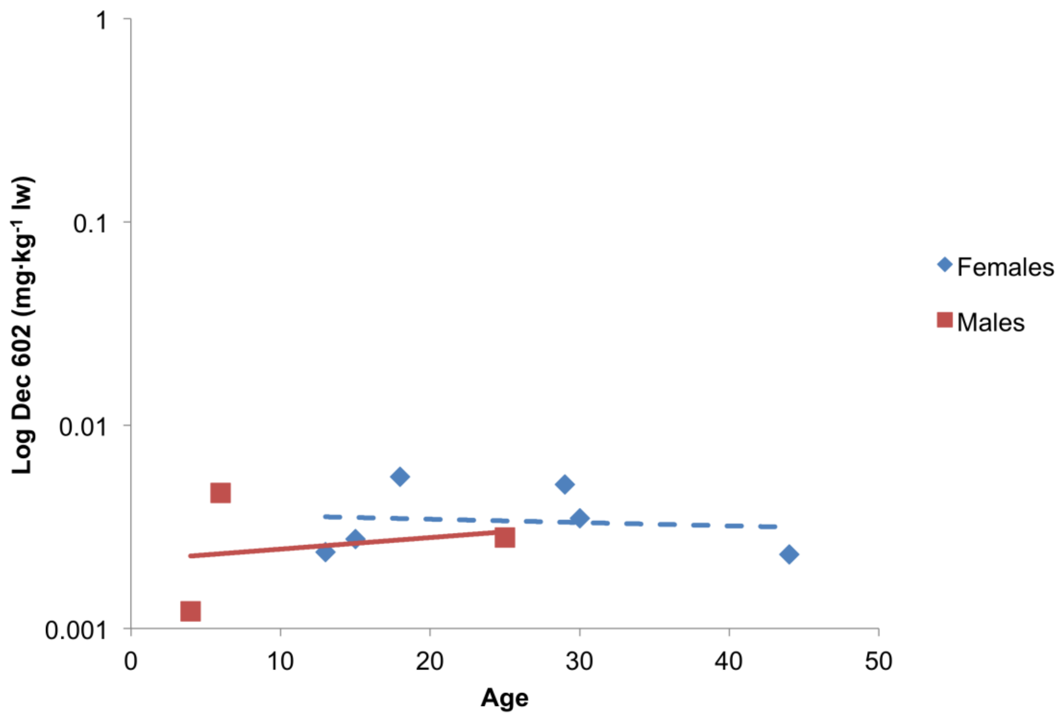

Figure 4.39 **The log-transformed concentrations of Dechlorane 602 (Dec 602; mg·kg^{-1} lw) in southern resident killer whales as a function of the ages (in years) of killer whales sampled in 2015. Two linear regression analyses are shown, one for females (◆) and the other for males (■).**

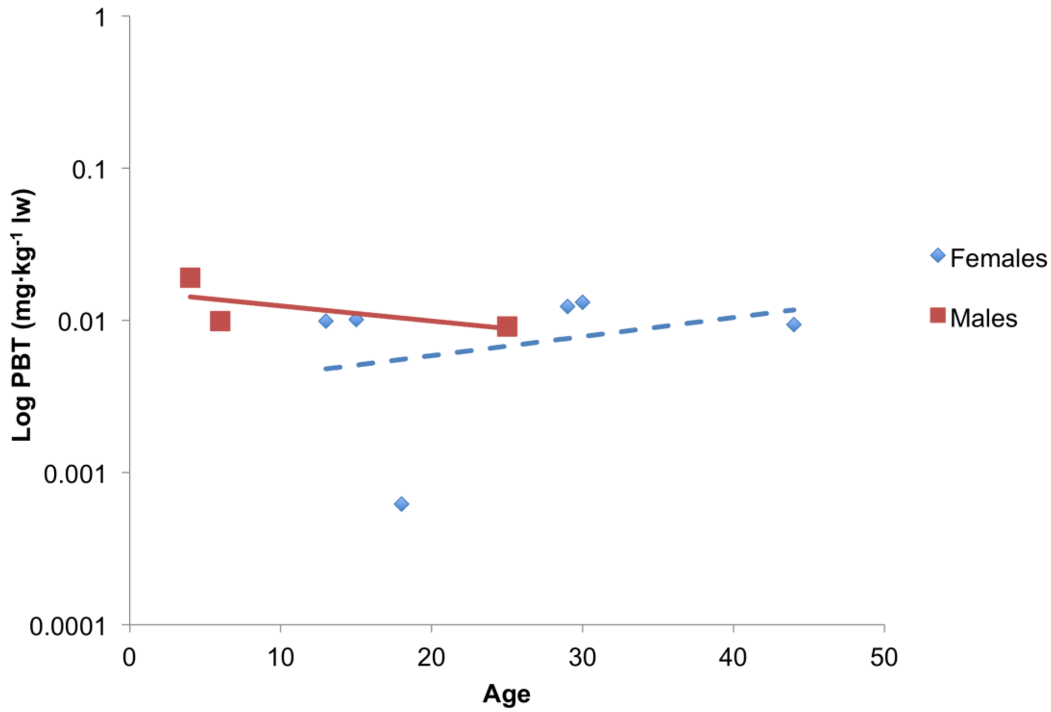

Figure 4.40 **The log-transformed concentrations of pentabromotoluene (PBT; mg·kg^{-1} lw) in southern resident killer whales as a function of the ages (in years) of killer whales sampled in 2015. Two linear regression analyses are shown, one for females (◆) and the other for males (■).**

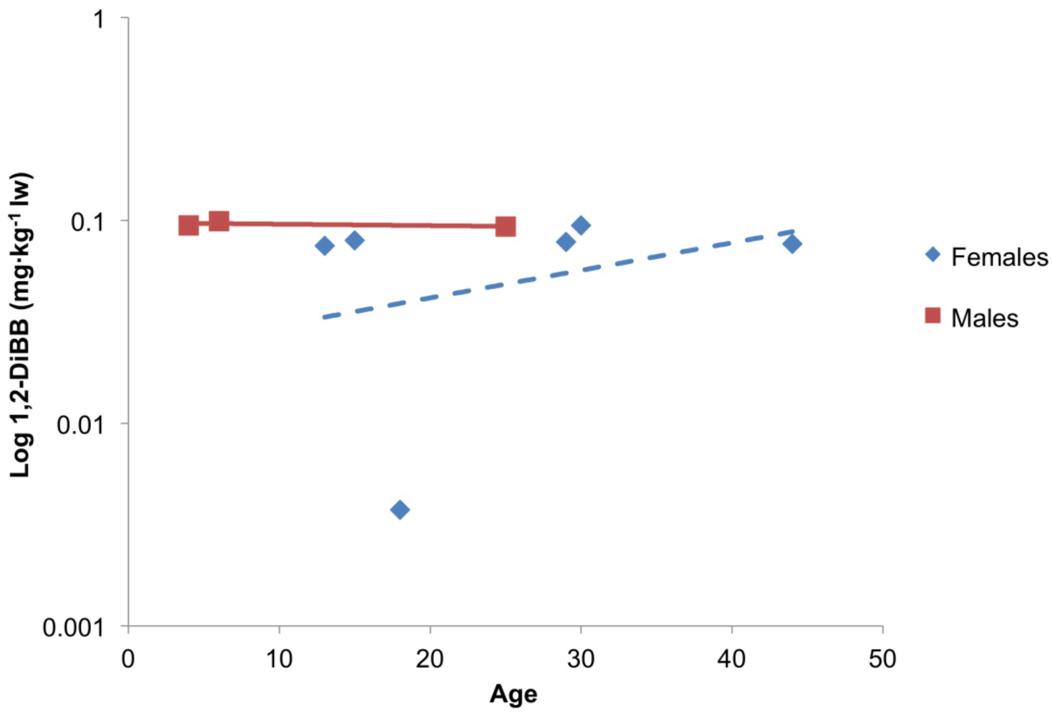

Figure 4.41 **The log-tranformed concentrations of 1,2-dibromobenzene (1,2-DiBB; mg·kg^{-1} lw) in southern resident killer whales as a function of the ages (in years) of killer whales sampled in 2015. Two linear regression analyses are shown, one for females (◆) and the other for males (■).**

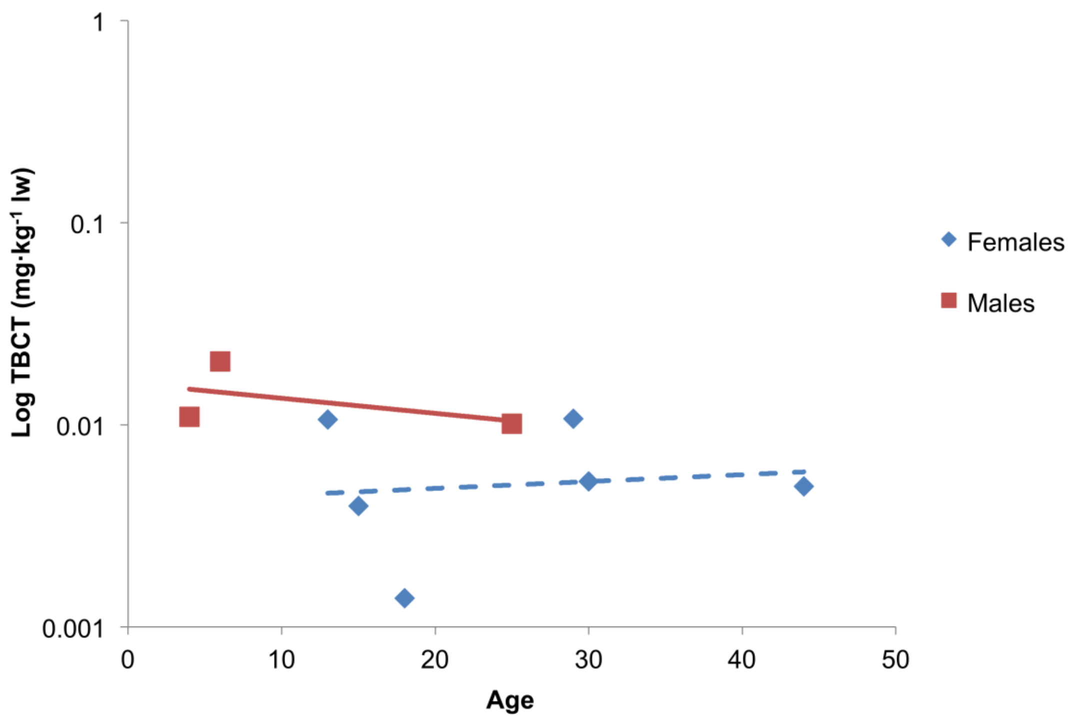

Figure 4.42 The log-transformed concentrations of tetrabromo-o-chlorotoluene (TBCT; mg·kg^{-1} lw) in southern resident killer whales as a function of the ages (in years) of killer whales sampled in 2015. Two linear regression analyses are shown, one for females (◆) and the other for males (■).

Table 4.16 Results of linear regression analyses for age versus concentrations of emerging contaminants in southern resident killer whales sampled in 2015. The emerging contaminants investigated included total hexabromocyclododecane isomers (ΣHBCDDs), Dechlorane, Dechlorane plus, Dechlorane 602 (Dec 602), 1,2-dibromobenzene (1,2-DiBB), pentabromotoluene (PBT) and tetrabromo-o-chlorotoluene (TBCT). Linear regression analyses were conducted separately for males and females.

Contaminant	Sex	Linear Regression	r^2	P-value
Dec 602	Female	Log Dec 602 = -2.43 - 0.00164*Age	1.36E-02	0.826
1,2-DiBB	Female	Log 1,2-DiBB = -1.65 + 0.0136*Age	8.61E-02	0.572
Dechlorane Plus	Female	Log Dechlorane Plus = -2.63 + 0.0151*Age	2.19E-01	0.349
Dechlorane	Female	Log Dechlorane = -1.10 - 0.00424*Age	4.92E-02	0.673
ΣHBCDDs	Female	Log ΣHBCDDs = -1.12 - 0.00474*Age	4.35E-01	0.154

Contaminant	Sex	Linear Regression	r^2	P-value
PBT	Female	Log PBT = -2.48 + 0.0125*Age	8.35E-02	0.579
TBCT	Female	Log TBCT = -2.38 + 0.00339*Age	1.49E-02	0.818
Dec 602	Male	Log Dec 602 = -2.67 + 0.00568*Age	5.02E-02	0.856
1,2-DiBB	Male	Log 1,2-DiBB = -1.01 - 0.000722*Age	3.29E-01	0.611
Dechlorane Plus	Male	Log Dechlorane Plus = -1.97 - 0.00455*Age	6.03E-01	0.434
Dechlorane	Male	Log Dechlorane = -0.960 - 0.0129*Age	2.09E-01	0.698
ΣHBCDDs	Male	Log ΣHBCDDs = -0.844 - 0.0163*Age	9.87E-01	0.0734
PBT	Male	Log PBT = -1.81 - 0.00992*Age	4.26E-01	0.548
TBCT	Male	Log TBCT = -1.79 - 0.00747*Age	2.63E-01	0.657

4.2.6. Linear Regression – Chinook Salmon

Linear regression analysis found no statistically significant relationship between the log-transformed concentrations of ΣPCBs measured in Chinook salmon and the year in which the salmon were sampled (Figure 4.43; Table 4.17). Linear regression analysis of concentrations of ΣPBDEs in Chinook salmon in relation to sample year was not conducted because there was an insufficient sample size (Figure 4.44; only n = 1 in 2002 and then n = 6 in 2014).

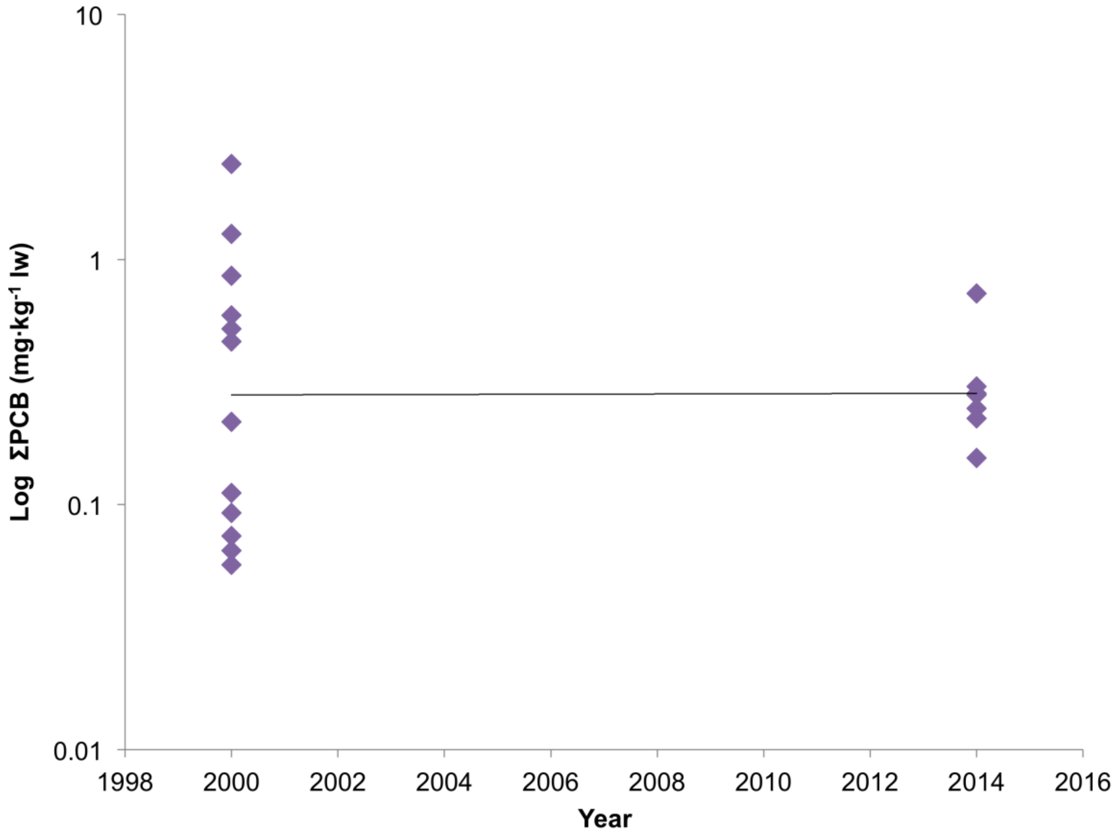

Figure 4.43 **Log-transformed concentrations of total polychlorinated biphenyls (ΣPCBs; mg·kg^{-1} lw) in Chinook salmon plotted against the years in which fish were sampled.**

Table 4.17 **Linear regression analysis of the log-transformed concentrations of total polychlorinated biphenyls (ΣPCBs) in Chinook salmon versus the years in which fish were sampled for ΣPCBs.**

Contaminant	Animal	Linear Regression	r^2	P-value
ΣPCBs	Chinook salmon	Log (ΣPCB) = -1.50 + 0.000475*Year	5.34E-05	0.976

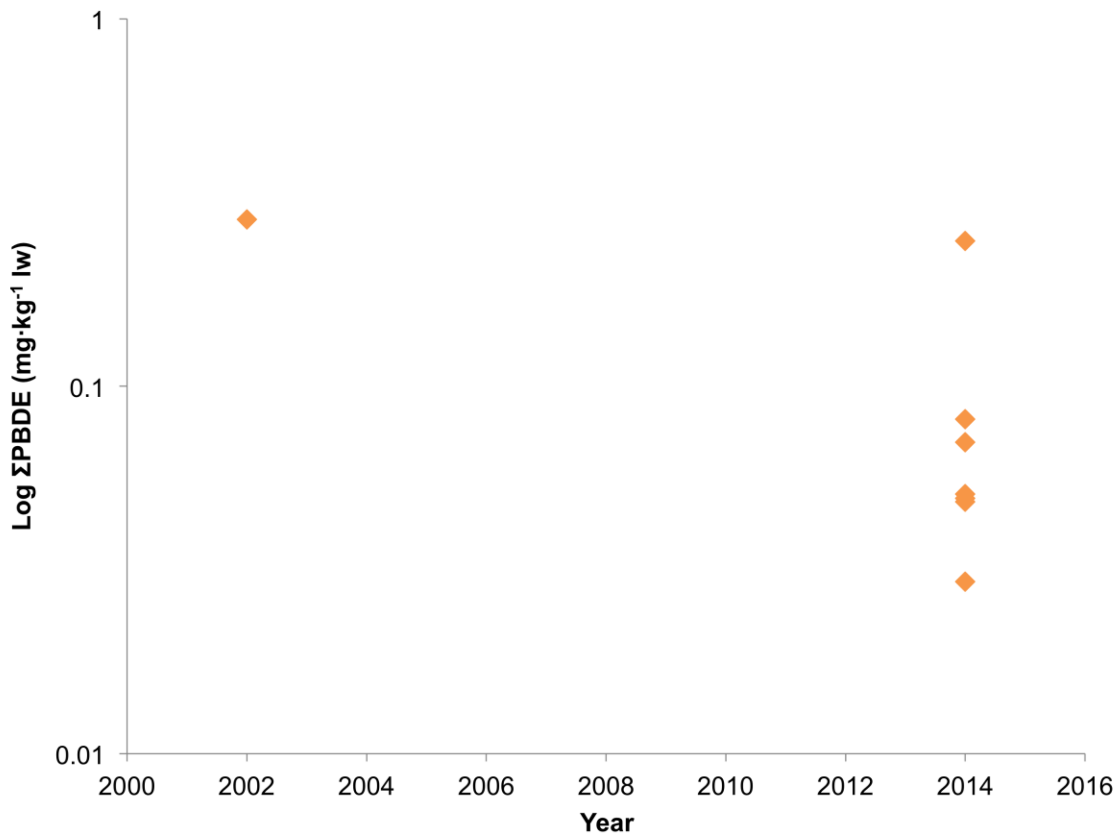

Figure 4.44 Log-transformed concentrations of total polybrominated diphenyl ethers (ΣPBDEs) measured in Chinook salmon samples plotted against the years in which fish were sampled.

Concentrations of ΣHBCDDs and other emerging flame-retardants were analyzed in new Chinook salmon samples collected in 2014 (Appendix A). The geometric mean for ΣHBCDDs was 7.43 $\times 10^{-3}$ mg·kg^{-1} lw, with upper and lower standard deviations of 1.18$\times 10^{-2}$ and 4.68$\times 10^{-3}$ mg·kg^{-1} lw, respectively. All chemicals listed in Appendix A were tested and no other emerging flame-retardants were detected in the 2014 Chinook salmon samples.

There was a 14-year and a 12-year gap in the data for which no concentrations of ΣPCBs and ΣPBDEs in Chinook salmon were available (Figures 4.43 and 4.44, respectively). More data concerning concentrations of ΣPCBs and ΣPBDEs in Chinook salmon are required to better understand how diet can effect present and future contaminant concentrations in northern and southern resident killer whales.

Hickie et al.[20] estimated the future trajectory of the concentration of PCBs in the salmon diet of northern and southern resident killer whales from the year 2000 to 2030. Two scenarios were analyzed. In the first, concentrations of PCBs in the salmon diet of killer whales remained the same (steady-state PCB exposure), causing concentrations of PCBs in killer whales in 2000 to be similar to those in 2030. The second scenario showed an environmental half-life of 30 years for PCBs in the salmon diet, resulting in an outcome where the concentrations of PCBs declined steadily in killer whales over the 30-year period. The concentrations of PCBs in Chinook salmon have not changed from 2000 to 2014, and the concentrations of PCBs in killer whales have also not changed significantly over the same time period. This would suggest that Hickie et al.'s[20] model that tested for steady-state exposure was a more accurate representation of the concentrations killer whales are experiencing in reality. If future concentrations of PCBs and PBDEs in Chinook salmon can be measured, these modelled trajectories could be tested for their accuracy.

4.2.7. ΣPBDE/ΣPCB Ratio Comparisons

Killer Whales – Ratio Comparisons

To eliminate various possible confounding factors such as age, ratios of the emerging pollutants to the legacy pollutant, PCBs, were calculated for killer whales within the same sample year. Only killer whales sampled for both ΣPCBs and ΣPBDEs in the same sample year were considered for these calculations. A ratio below the value of one was considered to indicate that PCBs were the main contaminant of concern within the comparison.

The log-transformed ratios of ΣPBDEs/ΣPCBs in killer whales increased significantly over time (Figure 4.45; SE = 0.0110, p = 0.00310). Results of the linear regression analysis of the ΣPBDEs/ΣPCBs for all killer whales are presented in Table 4.18 (see Appendix H for individual killer whale ΣPBDEs/ΣPCBs ratios).

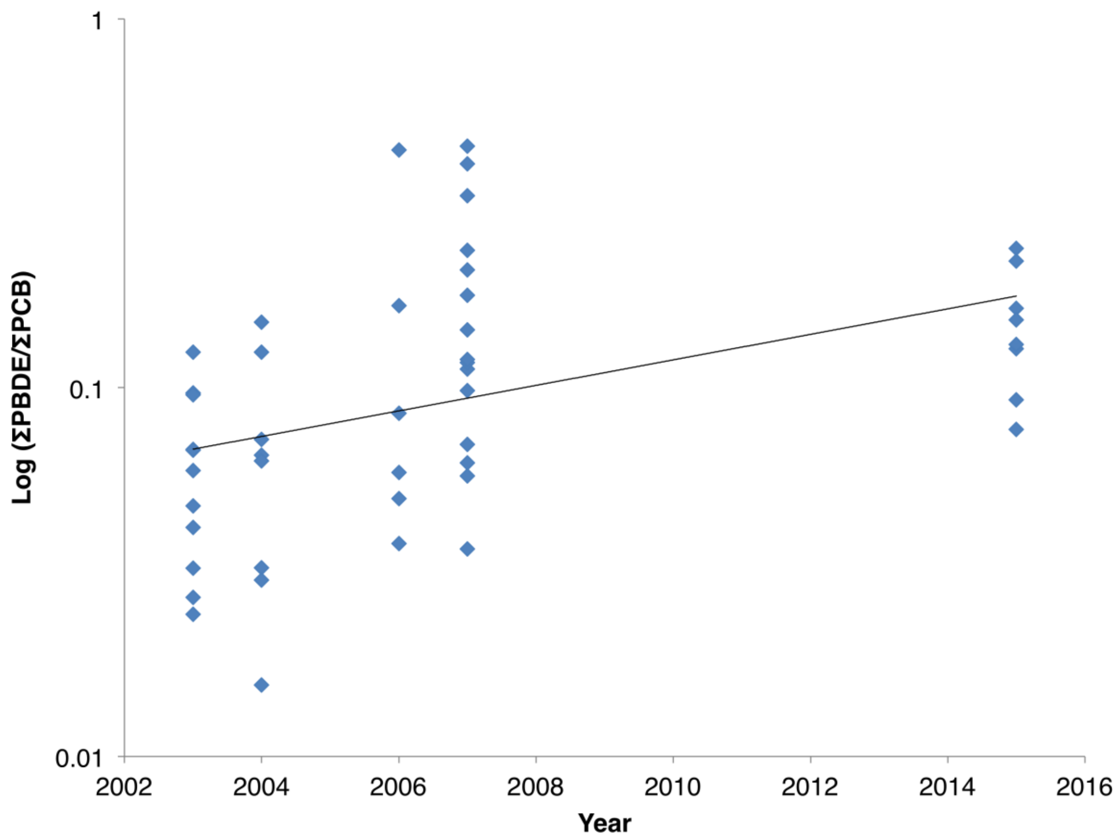

Figure 4.45 The log-transformed ratio of total polybrominated diphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in killer whales plotted against sample year. The solid line represents the linear relationship between the ΣPBDEs/ΣPCBs ratio and time.

Table 4.18 Results of the linear regression analysis of the log-transformed ratio of total polybrominated diphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) versus time for all killer whales.

Linear Regression	r^2	P-value	n
Log (ΣPBDE/ΣPCB) = -70.5 + 0.0346*Year	0.172	0.00310	48

The significant increase in the ratio of ΣPBDEs/ΣPCBs in killer whales over time showed that ΣPBDEs are becoming a contaminant of greater concern over time. All ΣPBDEs/ΣPCBs values were below one (see Appendix H), however, meaning that ΣPBDEs have not yet surpassed ΣPCBs as the main contaminant of concern.

The ratios of the log-transformed concentrations of ΣPBDEs/ΣPCBs in killer whales were plotted against sampling year, separately by sex and ecotype (Figures 4.46

– 4.51). Outcomes of the linear regression analysis of the log-transformed concentrations of ΣPBDEs/ΣPCBs in relation to sampling year are presented separately by sex and ecotype in Table 4.19. Linear regression analyses were not conducted for male Bigg's killer whales (n = 2 in 2003, n = 1 in 2007) or female southern resident killer whales (n = 1 in 2006, n = 6 in 2015) due to insufficient data. No significant changes in the log-transformed concentrations of ΣPBDEs/ΣPCBs were found to occur in relation to sampling year, regardless of sex or ecotype (see Table 4.19).

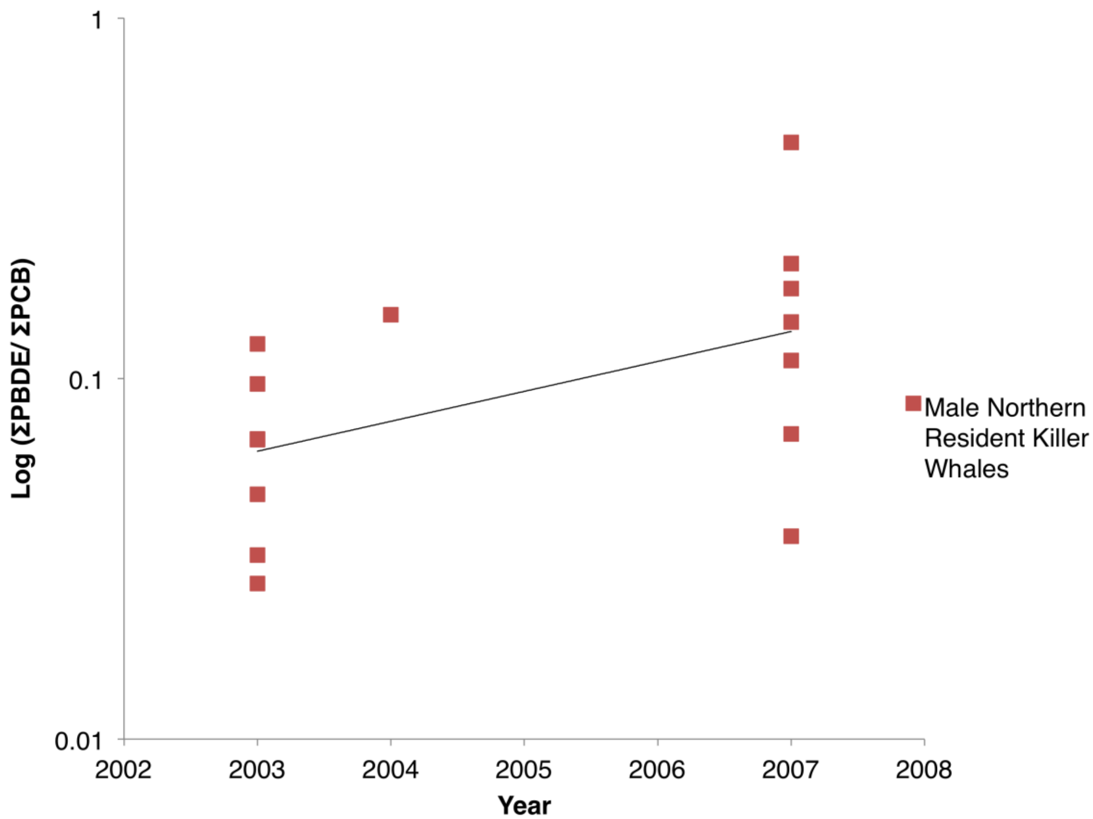

Figure 4.46 **Log-transformed ratios of total polybrominated biphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in male northern resident killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of the ratio of ΣPBDEs/ΣPCBs over time.**

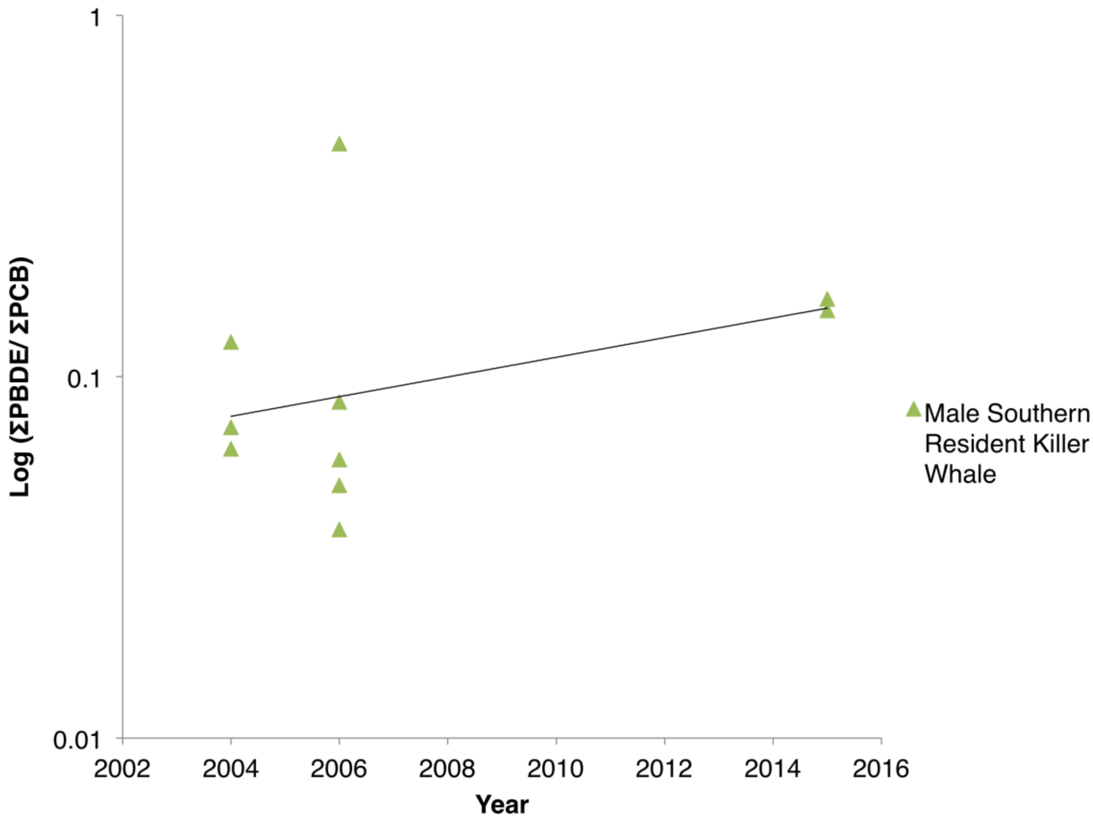

Figure 4.47 **Log-transformed ratios of total polybrominated biphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in male southern resident killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of the ratio of ΣPBDEs/ΣPCBs over time.**

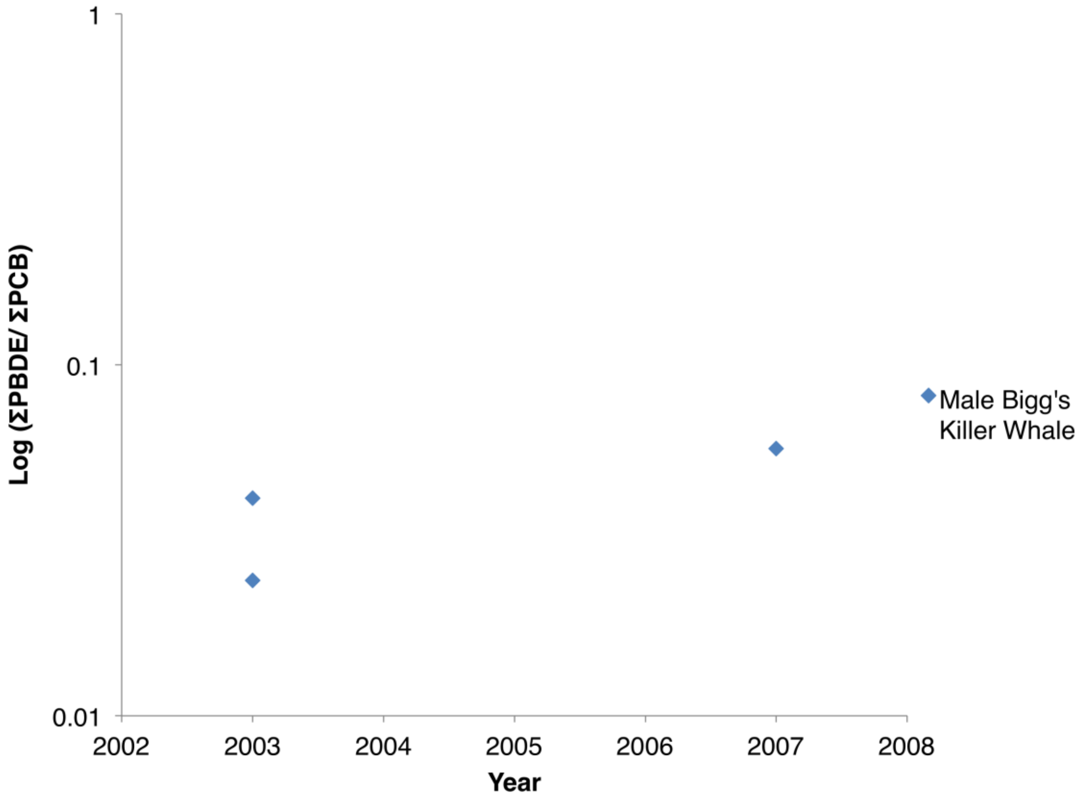

Figure 4.48 **Log-transformed ratios of total polybrominated biphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in male Bigg's killer whales plotted against the year in which each whale was sampled.**

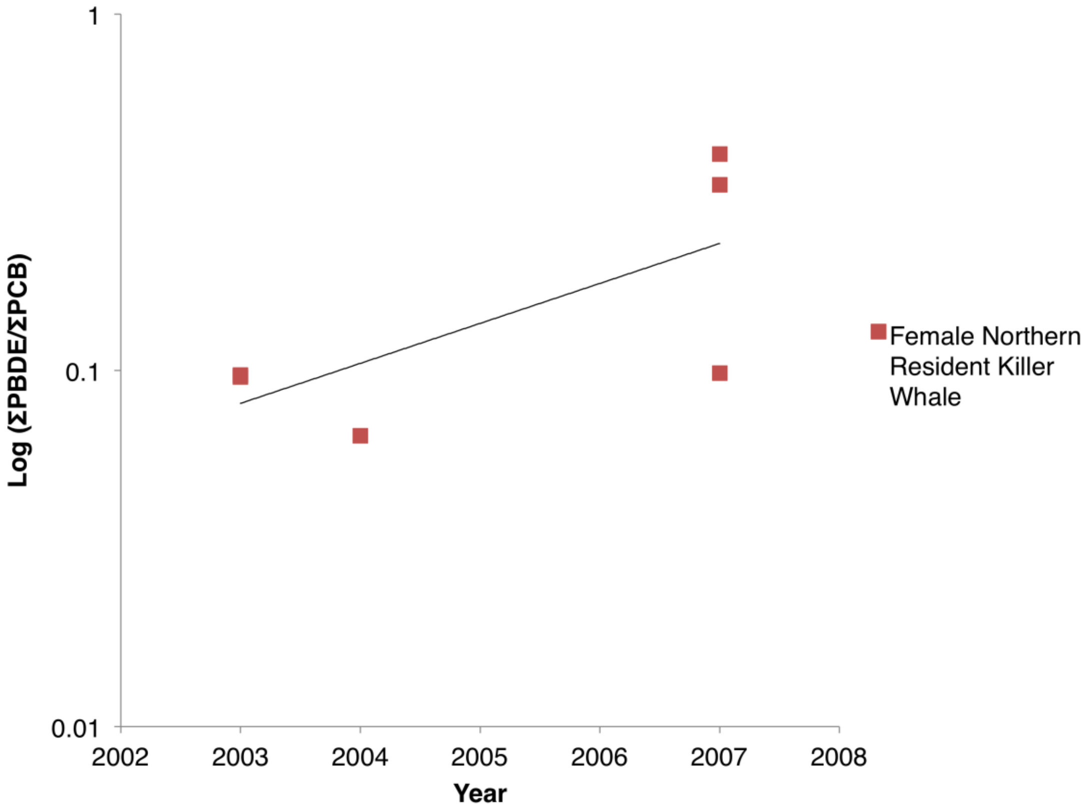

Figure 4.49 **Log-transformed ratios of total polybrominated biphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in female northern resident killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of the ratio of ΣPBDEs/ΣPCBs over time.**

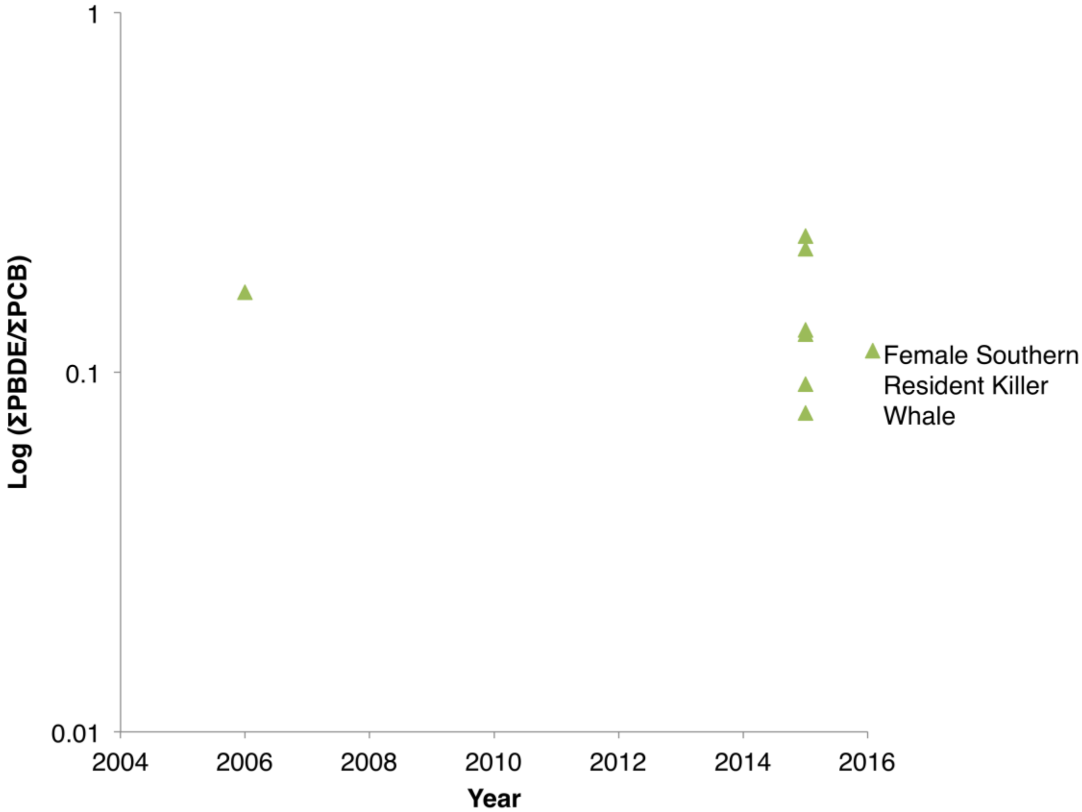

Figure 4.50 Log-transformed ratios of total polybrominated biphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in female southern resident killer whales plotted against the year in which each whale was sampled.

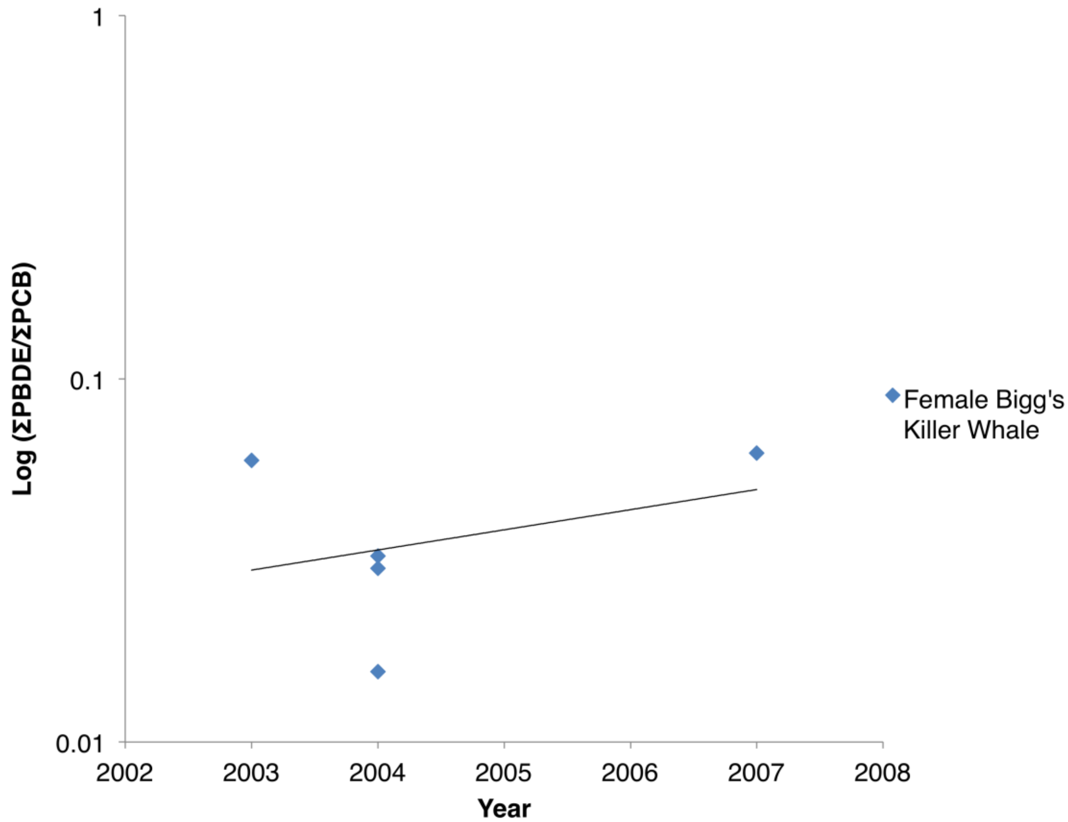

Figure 4.51 **Log-transformed ratios of total polybrominated biphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in female Bigg's killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of the ratio of ΣPBDEs/ΣPCBs over time.**

Table 4.19 **Results of linear regression analyses of the log-transformed ratio of total polybrominated biphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) over time. Linear regression analyses were conducted separately by sex and by ecotype – northern resident (NR), southern resident (SR) and Bigg's. Due to insufficient data, linear regressions were not conducted for male Bigg's and female SR killer whales.**

Sex	Ecotype	Linear Regression	r^2	P-value
Male	NR	Log (ΣPBDE/ΣPCB) = -168 + 0.0831*Year	0.250	0.0579
Male	SR	Log (ΣPBDE/ΣPCB) = -55.7 + 0.0272*Year	0.133	0.299
Female	NR	Log (ΣPBDE/ΣPCB) = -226 + 0.112*Year	0.488	0.123
Female	Bigg's	Log (ΣPBDE/ΣPCB) = -113 + 0.0555*Year	0.116	0.576

When the ratios of ΣPBDEs/ΣPCBs were split by ecotype and sex, no significant relationships between the ratios and sampling year were found. This could be because sample sizes were small when the dataset was separated by sex and ecotype. More data are needed to better understand if PBDEs are truly surpassing PCBs as the main contaminant of concern.

The log-transformed ratios of emerging pollutants/ΣPCBs for all individual killer whales sampled in 2015 are shown in Figure 4.52. The emerging pollutants included ΣHBCDDs, Dechlorane, DP, Dec 602, PBT, 1,2-DiBB, and TBCT. The individual ratios calculated for each of the different emerging pollutants/ΣPCBs are given in Table 4.20. The log-transformed geometric means of each emerging pollutant/ΣPCBs ratio are depicted in Figure 4.53, and corresponding mean values for each ratio are presented in Table 4.21.

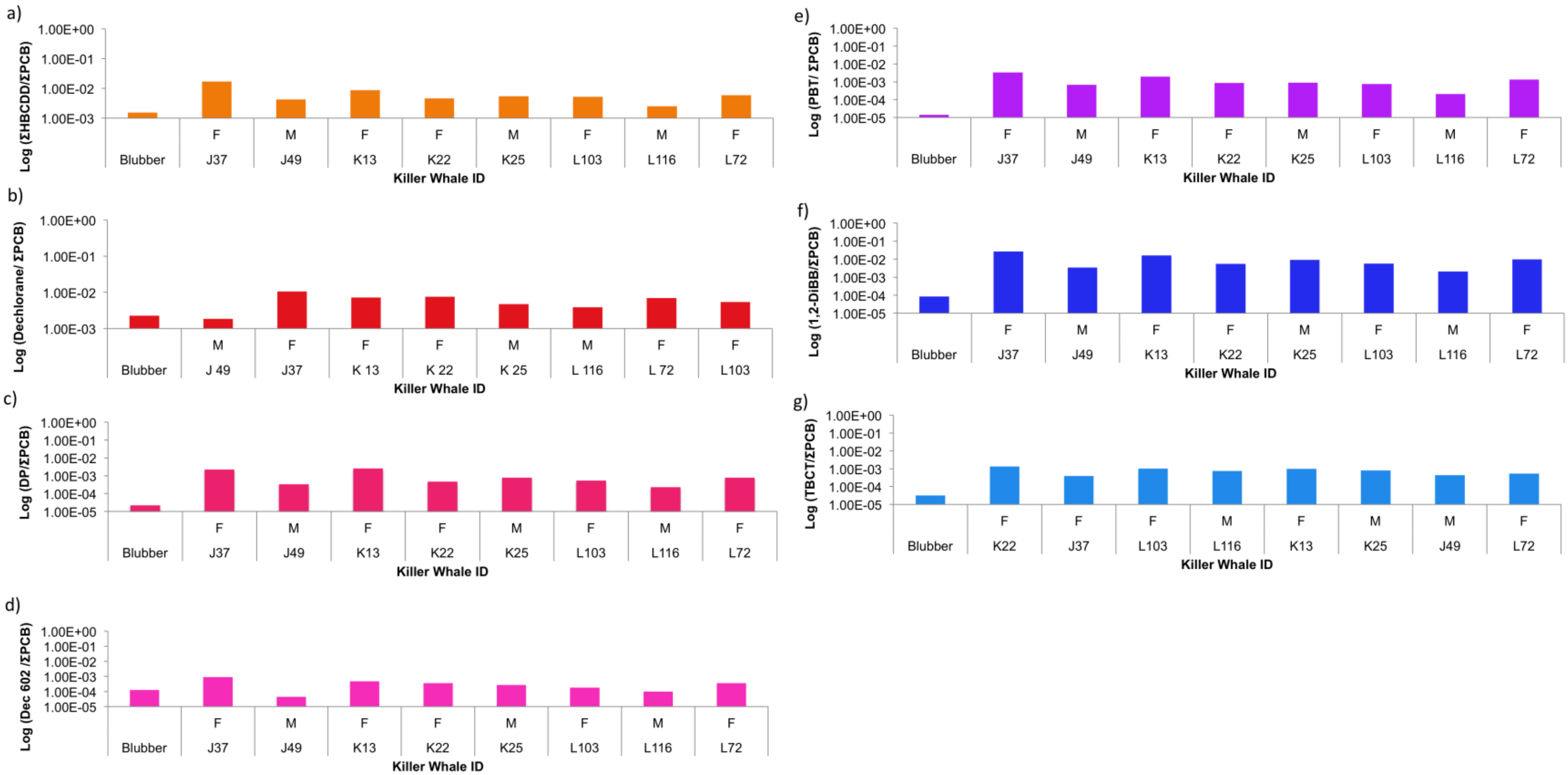

Figure 4.52 **Bar graphs showing the log-transformed ratios of emerging pollutants to total polychlorinated biphenyls (ΣPCBs) for each individual killer whale sampled in 2015. The ratios presented include: a) total isomers of hexabromocyclododecane (ΣHBCDDs) to ΣPCBs; b) Dechlorane to ΣPCBs; c) Dechlorane Plus (DP) to ΣPCBs; d) Dechlorane 602 (Dec 602) to ΣPCBs; e) pentabromotoluene (PBT) to ΣPCBs; f) 1,2-dibromobenzene (1,2-DiBB) to ΣPCBs; and g) tetrabromo-o-chlorotoluene (TBCT) to ΣPCBs.**

Table 4.20 **The ratios of seven emerging pollutants in comparison to total polychlorinated biphenyls (ΣPCBs) in individual killer whales sampled in 2015. Emerging pollutants were hexabromocyclododecane (HBCDD), Dechlorane, Dechlorane Plus (DP), Dechlorane 602 (Dec 602), pentabromotoluene (PBT), 1,2-dibromobenzene (1,2-DiBB) and tetrabromo-o-chlorotoluene (TBCT).**

Killer Whale ID	Age	Sex	HBCDD/ ΣPCB	Dechlorane/ ΣPCB	DP/ΣPCB	Dec 602/ ΣPCB	PBT/ ΣPCB	TBCT/ ΣPCB	1,2-DiBB/ ΣPCB
Blubber	-	-	1.55E-03	2.26E-03	2.22E-05	1.27E-04	1.41E-05	3.14E-05	8.46E-05
K22	15	F	1.71E-02	1.86E-03	2.25E-03	9.16E-04	3.36E-03	1.32E-03	2.65E-02
J37	4	F	4.24E-03	1.06E-02	3.32E-04	4.38E-05	6.88E-04	3.94E-04	3.40E-03
L103	44	F	8.75E-03	7.22E-03	2.52E-03	4.80E-04	1.94E-03	1.03E-03	1.59E-02
L116	29	M	4.66E-03	7.62E-03	4.69E-04	3.59E-04	8.67E-04	7.54E-04	5.50E-03
K13	25	F	5.41E-03	4.75E-03	7.80E-04	2.72E-04	8.84E-04	9.82E-04	9.05E-03
K25	13	M	5.19E-03	3.89E-03	5.36E-04	1.79E-04	7.42E-04	7.98E-04	5.62E-03
J49	6	M	2.53E-03	6.90E-03	2.31E-04	9.78E-05	2.08E-04	4.32E-04	2.09E-03
L72	30	F	5.93E-03	5.41E-03	7.68E-04	3.61E-04	1.36E-03	5.42E-04	9.77E-03

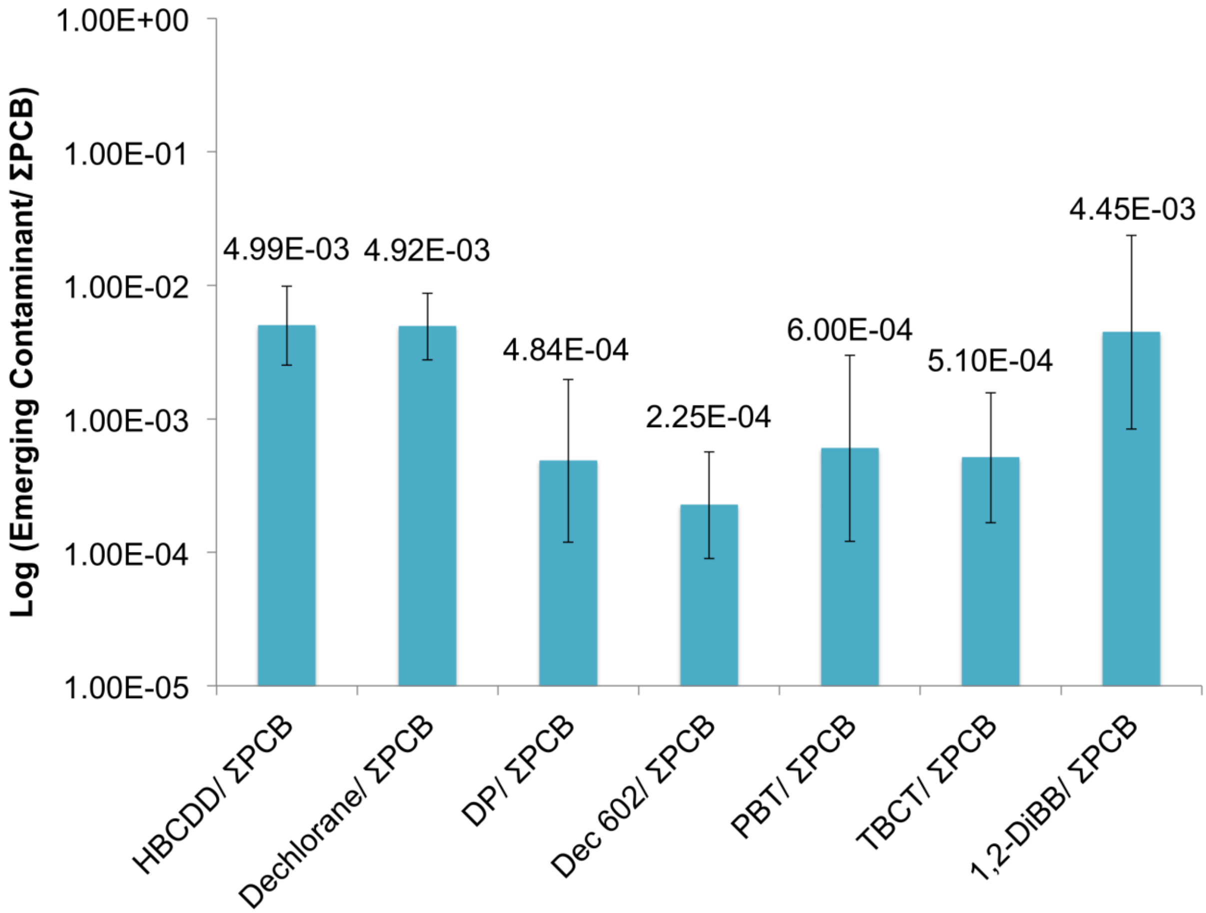

Figure 4.53 **Bar graph depicting the geometric means of the log-transformed ratios of emerging pollutants to total polychlorinated biphenyls (ΣPCBs) in killer whales sampled in 2015. Emerging pollutants were total isomers of hexabromocyclododecane (ΣHBCDDs), Dechlorane, Dechlorane Plus (DP), Dechlorane 602 (Dec 602), pentabromotoluene (PBT), 1,2-dibromobenzene (1,2-DiBB) and tetrabromo-o-chlorotoluene (TBCT). Error bars represent the standard deviation around each mean.**

Table 4.21 Geometric mean values for the ratios of emerging pollutants to total polychlorinated biphenyls (ΣPCBs) in all killer whales sampled in 2015. Emerging pollutants were hexabromocyclododecane (HBCDD), Dechlorane, Dechlorane Plus (DP), Dechlorane 602 (Dec 602), pentabromotoluene (PBT), tetrabromo-o-chlorotoluene (TBCT) and 1,2-dibromobenzene (1,2-DiBB).

Contaminant Ratio	Geometric Mean Ratio	Upper Standard Deviation	Lower Standard Deviation
HBCDD/ΣPCB	4.99E-03	9.88E-03	2.52E-03
Dechlorane/ΣPCB	4.92E-03	8.74E-03	2.77E-03
DP/ΣPCB	4.84E-04	1.97E-03	1.19E-04
Dec 602/ΣPCB	2.25E-04	5.66E-04	8.98E-05
PBT/ΣPCB	6.00E-04	2.98E-03	1.21E-04
TBCT/ΣPCB	5.10E-04	1.56E-03	1.66E-04
1,2-DiBB/ΣPCB	4.45E-03	2.37E-02	8.37E-04

The sample labeled as ‘Blubber’, which was taken from a deceased whale in 2015, exhibited the lowest ratios to ΣPCBs for five of the emerging contaminants investigated: ΣHBCDDs, DP, PBT, TBCT, and 1,2-DiBB. It is possible that the deceased whale was J32, a female killer whale that would have been 18 years of age when deceased. This shows that a whale may have higher concentrations of PCBs than other contaminants in the blubber before death. The reasons for the death of this whale are unknown; however, the high concentrations of the legacy pollutant, PCBs, compared to emerging pollutants in this whale highlight that the life history exposure of an animal may affect the concentrations of pollutants observed in that animal, thus impacting its health.

When exploring the geometric mean ratios, the lowest ratio was Dec 602/ΣPCBs, whereas ΣHBCDDs/ΣPCBs had the highest geometric mean ratio (Figure 4.53). This may indicate that of the seven emerging pollutants explored for this data set, HBCDD may be more of a concern to the health of killer whales. This was the first time these contaminants have been measured in killer whales, however, and future data are needed to better understand if HBCDD isomers are a major concern to killer whales. Overall, all geometric mean ratios were below one, showing that ΣPCBs were present in the highest concentrations and are thus of greater concern to the health of killer whales.

Killer Whales – Ratios of ΣPCBs and ΣPBDEs to Toxicity Reference Values

The ratios of ΣPCBs/TRV and ΣPBDEs/TRV were compared, using an average marine mammal TRV for ΣPCBs, calculated to be 1.5 $mg \cdot kg^{-1}$ lw, and a TRV for ΣPBDEs based on a study of grey seals (*Halichoerus grypus*), calculated as 1.5 $mg \cdot kg^{-1}$ lw.

Figure 4.54 – 4.56 shows the log-transformed ratios of the contaminants in female killer whales divided by the corresponding TRVs are shown in Figures 4.54 – 4.56, separated by ecotype. Linear regression analyses were also conducted for both ΣPCBs/TRV and ΣPBDEs/TRV.

A dashed-dotted line equalling one is included in Figures 4.54 – 4.56 to allow easier comparison of the concentrations/TRVs to one. If the concentration ratios were above one, the initial contaminant concentrations were more toxic than the TRV. Results of the linear regression of the log-transformed concentrations/TRVs for female killer whales in relation to sampling year are presented in Table 4.22.

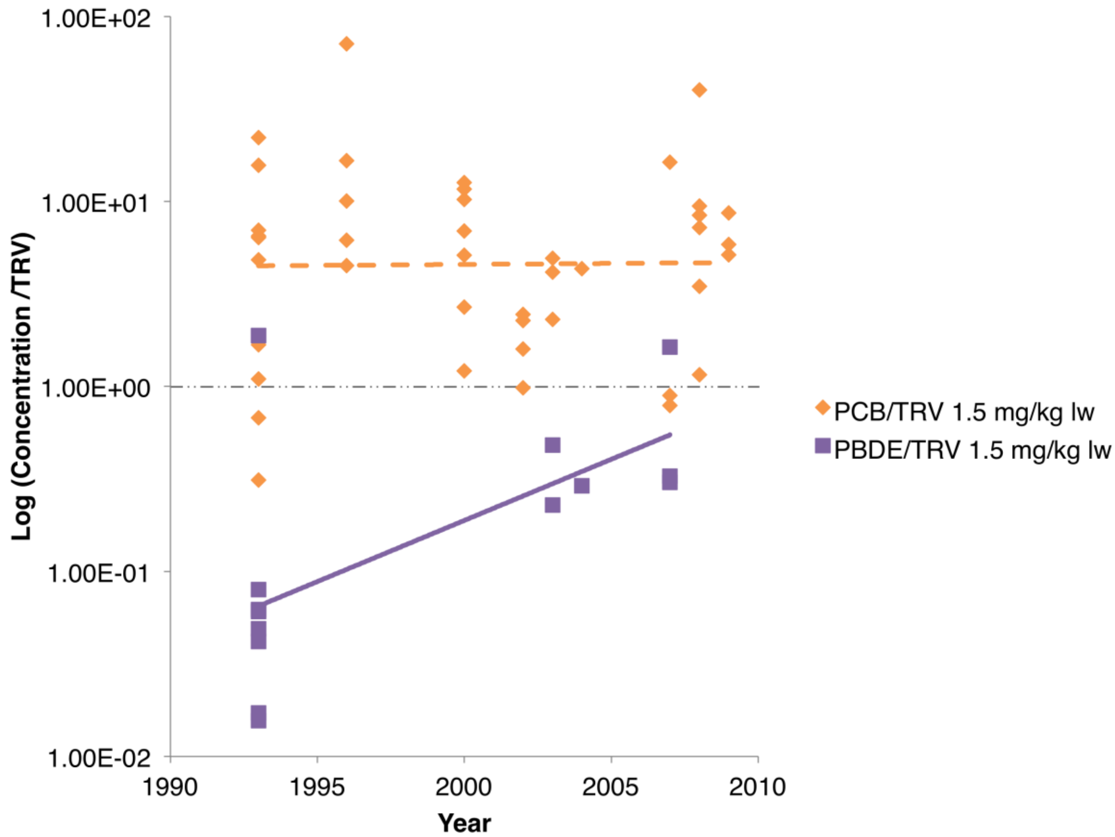

Figure 4.54 **The log-transformed ratios of concentrations of polychlorinated biphenyls (PCBs) to toxicity reference values (TRV), and concentrations of polybrominated diphenyl ethers (PBDEs) to TRV, for female northern resident killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of ΣPBDEs/TRV over time, and the dashed line gives the linear regression of ΣPCBs/TRV over time. The dashed-dotted line shows when the log-transformed contaminant concentration/TRV is equal to one.**

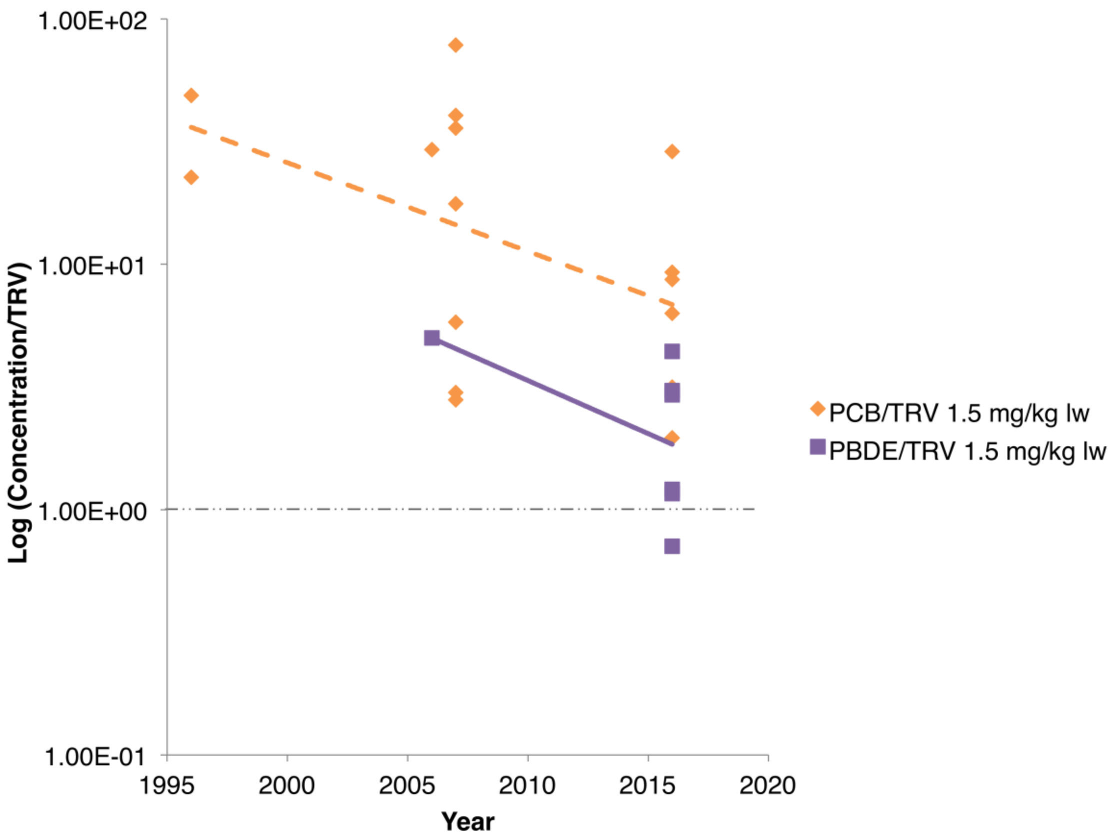

Figure 4.55 **The log-transformed ratios of concentrations of polychlorinated biphenyls (PCBs) to toxicity reference values (TRV), and concentrations of polybrominated diphenyl ethers (PBDEs) to TRV, for female southern resident killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of ΣPBDEs/TRV over time, and the dashed line gives the linear regression of ΣPCBs/TRV over time. The dashed-dotted line shows when the log-transformed contaminant concentration/TRV is equal to one.**

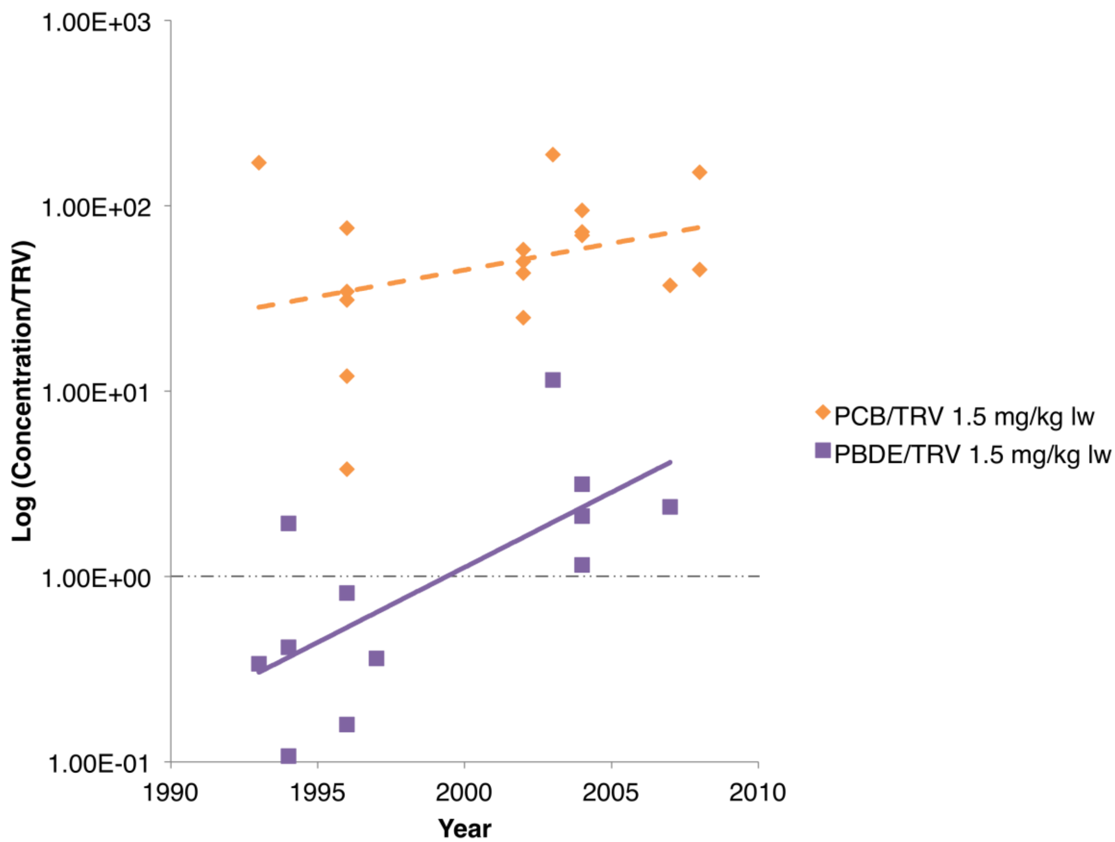

Figure 4.56 **The log-transformed ratios of concentrations of polychlorinated biphenyls (PCBs) to toxicity reference values (TRV), and concentrations of polybrominated diphenyl ethers (PBDEs) to TRV, for female Bigg's killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of ΣPBDEs/TRV over time, and the dashed line gives the linear regression of ΣPCBs/TRV over time. The dashed-dotted line shows when the log-transformed contaminant concentration/TRV is equal to one.**

Table 4.22 **Results of the linear regression analyses of the log-transformed ratios of concentrations of total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs) to toxicity reference values (TRV) over time in female killer whales. Analyses were conducted separately for each ecotype – southern resident (SR), northern resident (NR) and Bigg's.**

Ecotype	Sex	Contaminant	Linear Regression	r^2	P-value
SR	F	PCBs	Log (ΣPCBs/TRV) = 73.7 - 0.0361*Year	0.226	0.0627
NR	F	PCBs	Log (ΣPCBs/TRV) = -1.49 + 0.00107*Year	0.000158	0.937
Bigg's	F	PCBs	Log (ΣPCBs/TRV) = -55.7 + 0.0287*Year	0.104	0.206
SR	F	PBDEs	Log (ΣPBDEs/TRV) = 87.6 - 0.0433*Year	0.251	0.252
NR	F	PBDEs	Log (ΣPBDEs/TRV) = -1.33E+02+ 0.0661*Year	0.41	0.0137
Bigg's	F	PBDEs	Log (ΣPBDEs/TRV) = -1.62E+02 + 0.0809*Year	0.493	0.011

No significant relationships were found between the ratios of ΣPCBs/TRV and time for any female ecotypes (NR: SE = 0.0135, p = 0.937; SR: SE = 0.179, p = 0.0627; Bigg's: SE = 0.0217, p = 0.206). In contrast, the log-transformed ratios of ΣPBDEs/TRV were found to significantly increase over time for both female northern resident (SE = 0.0229, p = 0.0137) and Bigg's killer whales (SE = 0.0260, p = 0.0110). This indicates that the concentrations of PBDEs in both northern resident and Bigg's female killer whales may be exhibiting toxic effects, and could negatively affect these whales over time. This shows that PBDEs are becoming more toxic over time, whereas the toxicity of PCBs appears to have remained the same over time.

Based on linear regression, the ΣPCBs/TRV ratios for female northern resident killer whales were visually observed to be above one, whereas ΣPBDE/TRV ratios for northern resident female killer whales were below one. This indicates that the concentrations of ΣPCBs in female northern resident killer whales were greater than the TRV for those contaminants. Concentrations of ΣPBDEs, however, were not greater than the TRV for ΣPBDEs, suggesting that for northern resident female killer whales, ΣPCBs are present at a more toxic level than are ΣPBDEs. For female southern resident killer whales, both ΣPCBs/TRV and ΣPBDEs/TRV ratios were above one, meaning that both contaminants were present in toxic concentrations in these whales. Bigg's female killer whales exhibited ΣPCBs/TRV ratios that were above one, along with ΣPBDEs/TRV ratios that were above one in more recent sampling years (2004 and 2006), indicating that concentrations of ΣPCBs and recently-sampled concentrations of ΣPBDEs were considered toxic.

In past studies, such as those conducted by Ross et al.[1] and Rayne et al.[8], it was found that southern resident killer whales possessed more toxic concentrations of PCBs and PBDEs, respectively. The present study's findings are consistent with those past findings. Plausible reasons for highly toxic concentrations in southern resident killer whales could be due to inputs of pollutants in the habitat of southern resident killer whales[20].

The log-transformed ratios of ΣPCBs/TRV and ΣPBDEs/TRV in male killer whales were also analyzed in relation to time for all ecotypes (Figures 4.57 – 4.59). Outcomes of the linear regressions of both ΣPCBs/TRV and ΣPBDEs/TRV over the sample period for all ecotypes of male killer whales are given in Table 4.23.

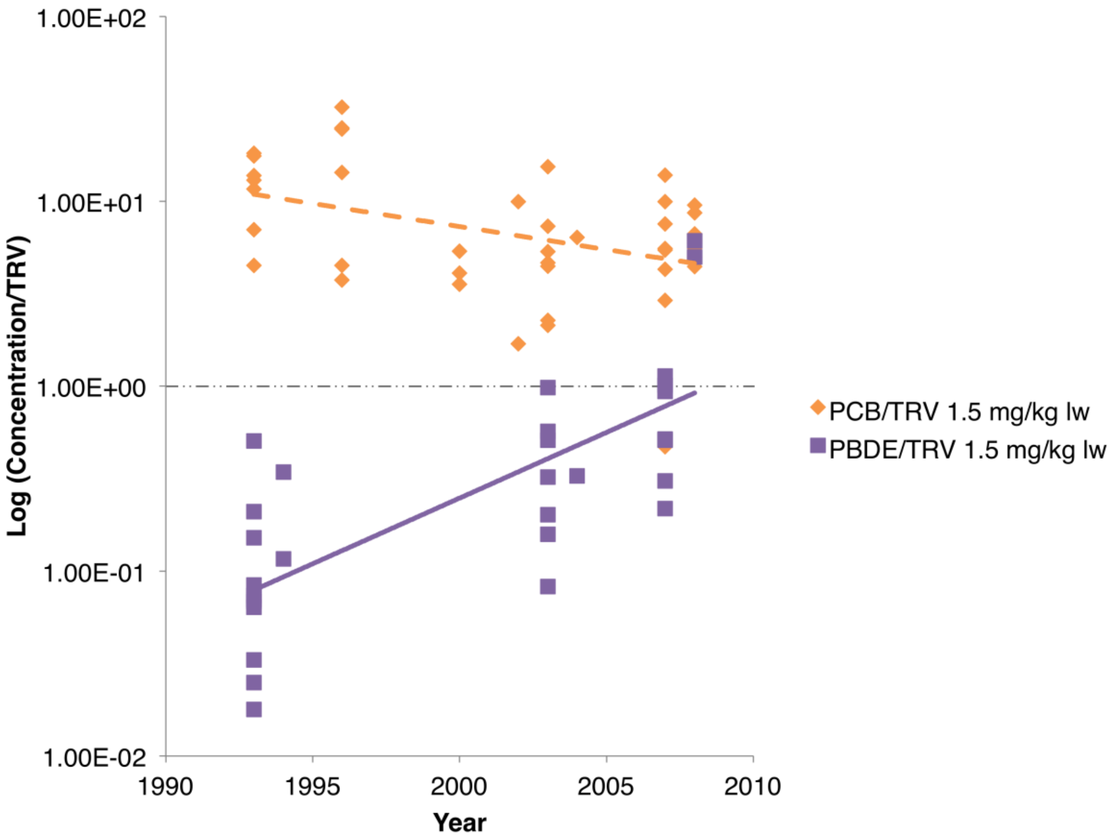

Figure 4.57 **The log-transformed ratios of concentrations of polychlorinated biphenyls (PCBs) to toxicity reference values (TRV), and concentrations of polybrominated diphenyl ethers (PBDEs) to TRV, for male northern resident killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of ΣPBDEs/TRV over time, and the dashed line gives the linear regression of ΣPCBs/TRV over time. The dashed-dotted line shows when the log-transformed contaminant concentration/TRV is equal to one.**

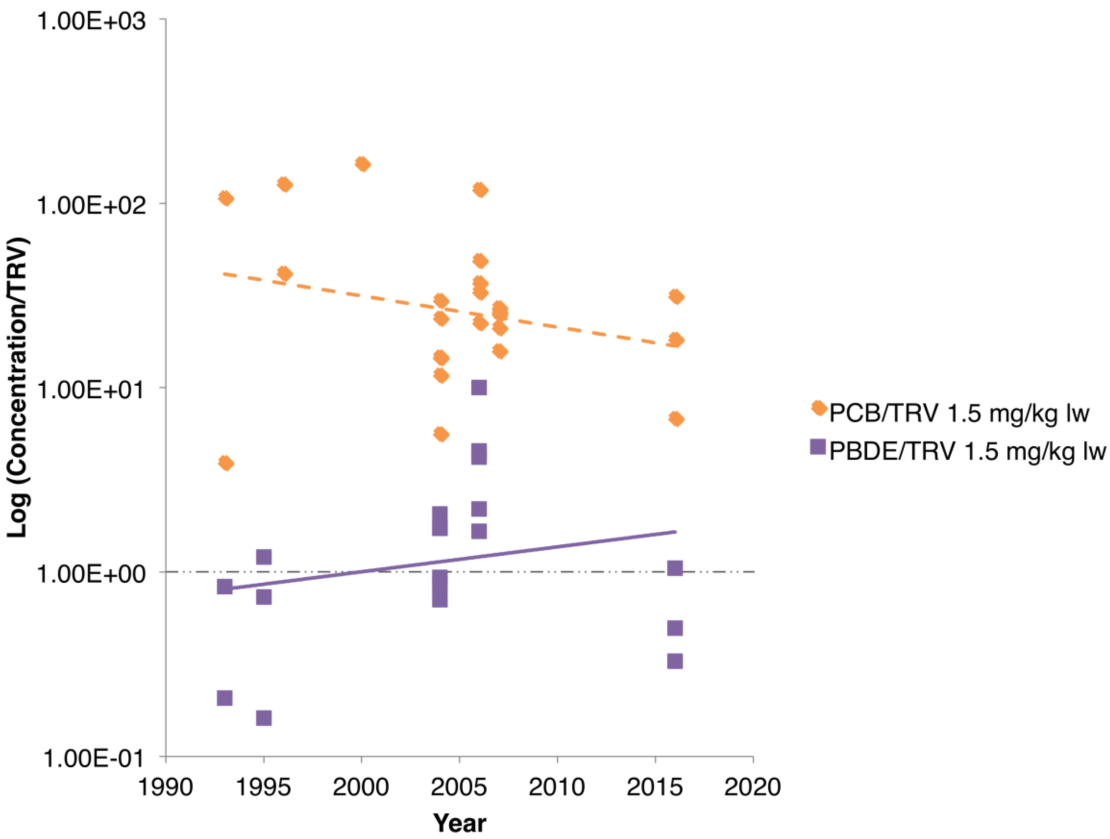

Figure 4.58 **The log-transformed ratios of concentrations of polychlorinated biphenyls (PCBs) to toxicity reference values (TRV), and concentrations of polybrominated diphenyl ethers (PBDEs) to TRV, for male southern resident killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of ΣPBDEs/TRV over time, and the dashed line gives the linear regression of ΣPCBs/TRV over time. The dashed-dotted line shows when the log-transformed contaminant concentration/TRV is equal to one.**

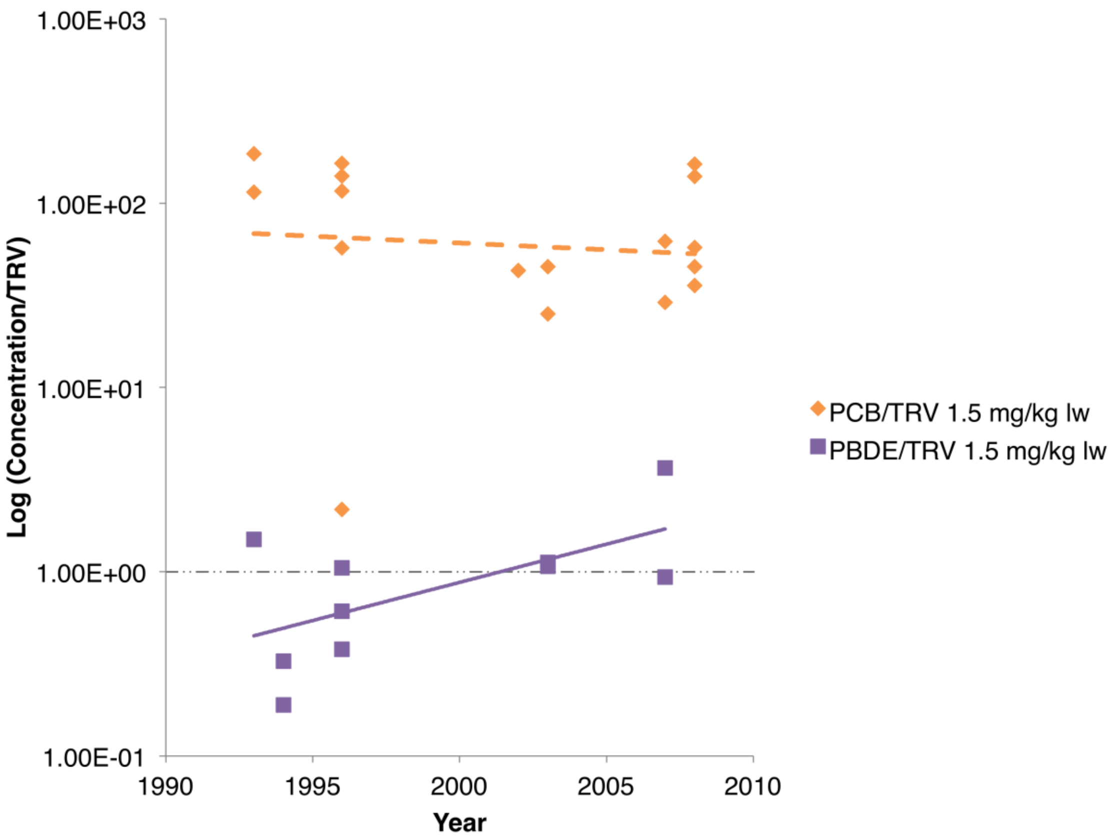

Figure 4.59 **The log-transformed ratios of concentrations of polychlorinated biphenyls (PCBs) to toxicity reference values (TRV), and concentrations of polybrominated diphenyl ethers (PBDEs) to TRV, for male Bigg's killer whales plotted against the year in which each whale was sampled. The solid line represents the linear regression of ΣPBDEs/TRV over time, and the dashed line gives the linear regression of ΣPCBs/TRV over time. The dashed-dotted line shows when the log-transformed contaminant concentration/TRV is equal to one.**

Table 4.23 **Results of the linear regression analyses of the log-transformed ratios of concentrations of total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs)to toxicity reference values (TRV) over time in male killer whales. Analyses were conducted separately for each ecotype – southern resident (SR), northern resident (NR) and Bigg's.**

Ecotype	Sex	Contaminant	Linear Regression	r^2	P-value
NR	M	PCBs	Log (PCBs/TRV) = 43.0 - 0.0211*Year	0.115	0.0241
SR	M	PCBs	Log (PCBs/TRV) = 35.4 - 0.0170*Year	0.0644	0.232
Bigg's	M	PCBs	Log (PCBs/TRV) = 14.7 - 0.00647*Year	0.00641	0.768
NR	M	PBDEs	Log (PBDEs/TRV) = -1.17E+02 + 0.0581*Year	0.509	<01.00E-04
SR	M	PBDEs	Log (PBDEs/TRV) = -27.0 + 0.0135*Year	0.0471	0.372
Bigg's	M	PBDEs	Log (PBDEs/TRV) = -82.9 + 0.0414*Year	0.382	0.057

A significant decreasing relationship was found between the log-transformed ratios of ΣPCBs/TRV and sample period for male northern resident killer whales (SE = 9.01E-03, $p = 0.00241$), whereas ΣPBDEs/TRV significantly increased over the sampling period (SE = 1.12E-03, $p < 0.00100$). This indicates that concentrations of ΣPCBs are reaching less toxic levels over time, whereas ΣPBDEs are increasing to greater levels of toxicity over time in male northern resident killer whales.

No significant relationships were found between ΣPCBs/TRV and time, or between ΣPBDEs/TRV and time, in male southern resident or Bigg's killer whales. This could be due to differences in the diets of Bigg's and resident killer whales. Another possibility could be due to insufficient data over the sample years. Regardless, more data are needed to determine if there are truly changes in the concentrations of PCBs and PBDEs in relation to TRV over time.

When comparing the ratios of contaminant concentrations/TRV to one, ΣPCBs/TRV were above one, showing that the concentrations of ΣPCBs in all three male killer whale ecotypes were above the TRV for ΣPCBs. For northern resident male killer whales, ΣPBDEs/TRV ratios were below one, indicating that concentrations of ΣPBDEs were not above the TRV for ΣPBDEs. In contrast, ΣPBDEs/TRV ratios were above one in more recent sampling year for southern resident and Bigg's male killer whales, indicating a similar outcome to that of female killer whales. Concentrations of ΣPCBs remain above TRVs, whereas ΣPBDEs are becoming more toxic in male killer whales. A study by Rayne et al. [8] supports the idea that PBDEs are reaching levels of greater toxicity in Salish Sea killer whales, and may therefore be a contaminant of concern for the health of killer whales.

It should be noted that there are possible limitations in using marine mammal TRVs for PCBs and PBDEs, as these marine mammal TRVs were derived from other marine mammals and not killer whales. Due to the lack of existing TRVs for killer whales, however, this is a widely accepted approach [7, 64]. It is assumed that the physiological response to the toxicity of contaminants is similar in all mammals, and this assumption allows the use of other marine mammal toxicity thresholds when assessing the health risks of killer whales [7, 64]. Furthermore, for the ratio calculations, equal toxicity of PCBs and PBDEs were assumed for the simplicity of the test, which was conducted to compare levels of PBDEs to those of the legacy pollutant, PCBs. It is possible that PCBs and PBDEs may not have equal toxicity and more research is needed to fully understand the toxicological comparisons of these two contaminants. The TRVs of both PCBs and PBDEs are of similar value, however, and thus a comparison based on the assumptions of equal toxicity is valid. Geometric mean concentrations of PCBs and PBDEs were also divided by their corresponding TRVs (see Appendix I).

Chinook Salmon

The main focus of the study was to compare the concentrations of detected contaminants, ΣPBDEs and ΣHBCDDs, to concentrations of ΣPCBs in Chinook salmon. The only year in which ΣPCBs, ΣPBDEs, and ΣHBCDDs were sampled and measured was 2014. Ratios of ΣPBDEs/ΣPCBs and ΣHBCDDs/ΣPCBs were thus calculated for 2014 and again, if the ratio was below one, it was deduced that PCBs were the main contaminant of concern in Chinook salmon.

Log-transformed ratios of ΣPBDEs/ΣPCBs for each individual Chinook salmon sampled in 2014 are shown in Figure 4.60, and corresponding log-transformed ratios of ΣHBCDDs/ΣPCBs are depicted in Figure 4.61. Individual ratio outcomes for both ΣPBDEs/ΣPCBs and ΣHBCDDs/ΣPCBs are presented in Table 4.24. The geometric means of ΣPBDEs/ΣPCBs and ΣHBCDDs/ΣPCBs are depicted in Figure 4.62 and reported in Table 4.25.

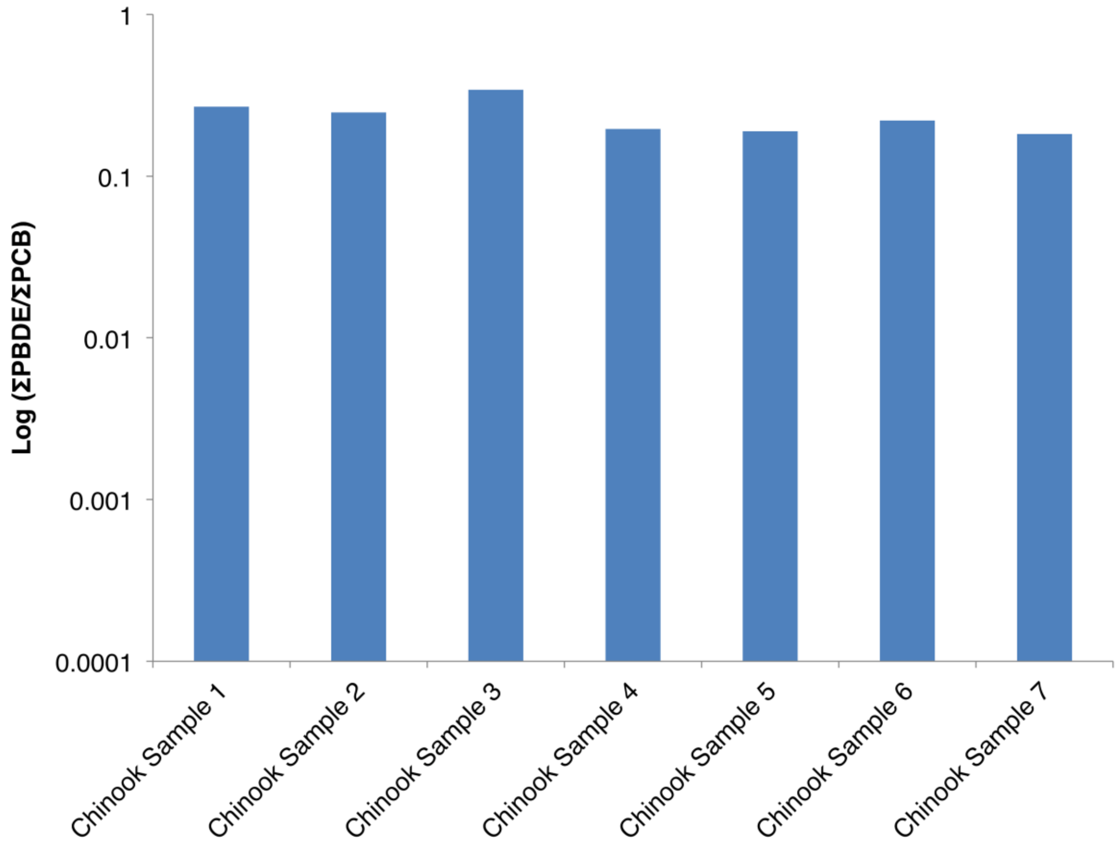

Figure 4.60 **Log-transformed ratios of total polybrominated diphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ ΣPCBs) in individual Chinook salmon sampled in 2014.**

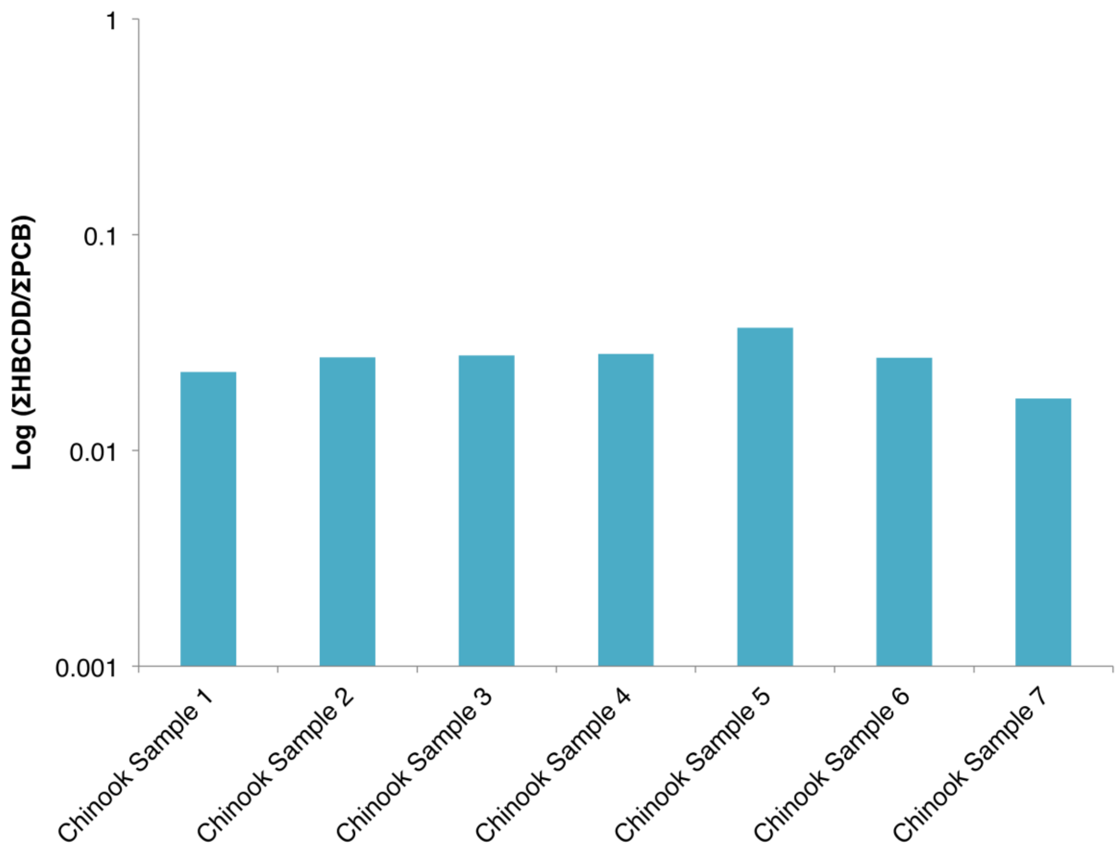

Figure 4.61 Log-transformed ratios of total hexabromocyclododecane isomers to total polychlorinated biphenyls (ΣHBCDDs/ΣPCBs) in individual Chinook salmon sampled in 2014.

Table 4.24 Ratios of total polybrominated diphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) and total hexabromocyclododecane isomers to total polychlorinated biphenyls (ΣHBCDDs/ΣPCBs) in individual Chinook salmon sampled in 2014.

Source	Year	Sample	ΣPBDEs/ ΣPCBs	ΣHBCDDs/ ΣPCBs
Present Study	2014	Chinook Sample 1	2.69E-01	2.31E-02
Present Study	2014	Chinook Sample 2	2.48E-01	2.69E-02
Present Study	2014	Chinook Sample 3	3.42E-01	2.75E-02
Present Study	2014	Chinook Sample 4	1.96E-01	2.81E-02
Present Study	2014	Chinook Sample 5	1.90E-01	3.70E-02
Present Study	2014	Chinook Sample 6	2.20E-01	2.69E-02
Present Study	2014	Chinook Sample 7	1.82E-01	1.74E-02

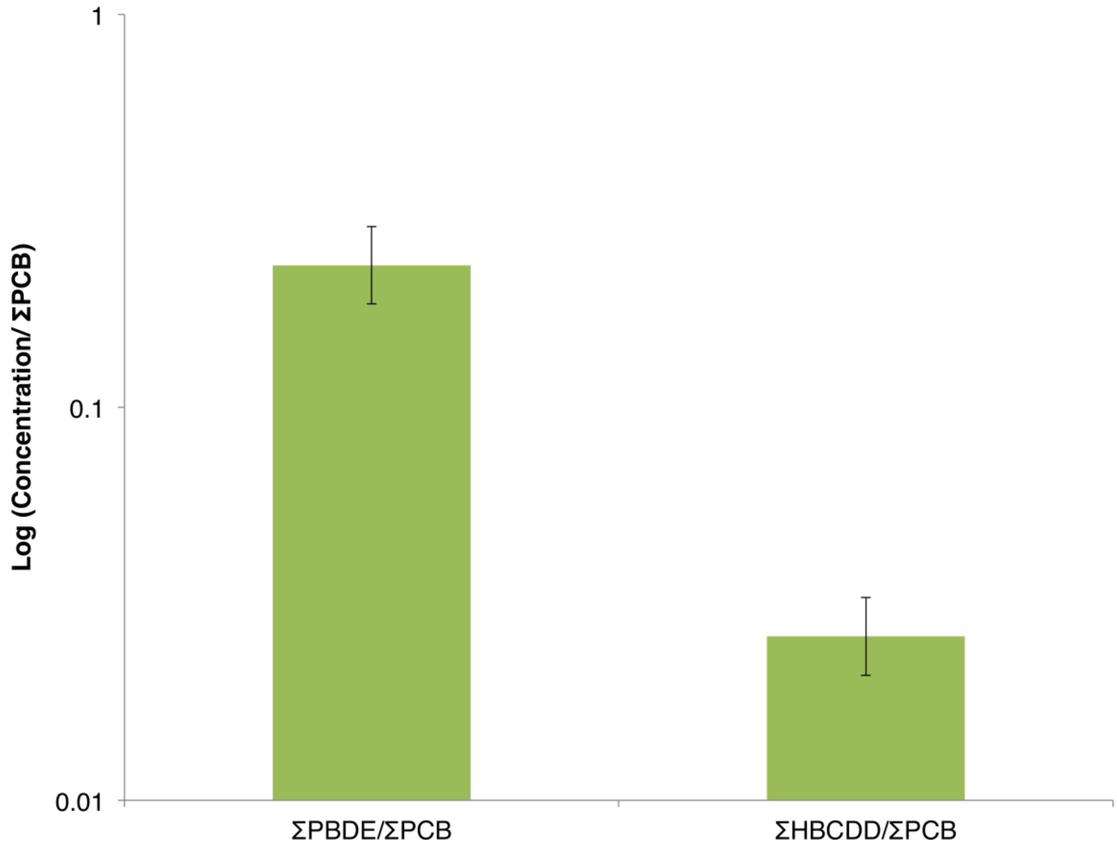

Figure 4.62 The geometric means of the ratios of total polybrominated diphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) and total hexabromocyclododecane isomers to total polychlorinated biphenyls (ΣHBCDDs/ΣPCBs) in Chinook salmon. Means are log-transformed. Error bars represent the standard deviation.

Table 4.25 The geometric means of the ratios of total polybrominated diphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) and total hexabromocyclododecane isomers to total polychlorinated biphenyls (ΣHBCDDs/ΣPCBs) in Chinook salmon, with their corresponding upper and lower standard deviations.

Ratio	Geometric Mean	Upper SD	Lower SD
ΣPBDE/ΣPCB	2.30E-01	2.88E-01	1.83E-01
ΣHBCDD/ΣPCB	2.61E-02	3.28E-02	2.08E-02

Outcomes for ΣPBDEs/ΣPCBs and ΣHBCDDs/ΣPCBs for every Chinook salmon sampled in 2014 were below the value of one, suggesting that ΣPCBs are present in greater concentrations in Chinook salmon than are ΣPBDEs and ΣHBCDDs. ΣPCBs were therefore considered to be the main pollutant of concern to the health of resident killer whales that consume Chinook salmon.

Concentrations of PCBs, PBDEs, and HBCDD in Chinook salmon were also divided by their corresponding Canadian wildlife diet guidelines. For PCBs, the CCME tissue residue guideline was originally expressed in TEQs. Hickie et al. [20] reworked the CCME PCB tissue residue guideline to 0.05 mg·kg^{-1} ww for PCBs to make the CCME PCB tissue residue guideline comparable to the manner in which concentrations of PCBs were typically measured. To do this, Hickie et al. [20] multiplied the CCME PCB tissue residue guideline against a regression of concentrations of PCBs measured in Chinook salmon data from a study by Cullon et al. [3].

For the current study, a new tissue residue guideline was calculated based on the concentrations of PCBs in Chinook salmon sampled for this study. The outcome determined to be 0.0688 mg·kg^{-1} ww, which is similar to the guideline suggested by Hickie al.[20]. The value of 0.0688 mg·kg^{-1} ww was therefore used for the concentration ratios explored in Chinook salmon in the present study.

For the PBDE FEQG, the guideline value was recalculated to be 0.00945 mg·kg^{-1} ww. This value was thus used to compare concentrations of ΣPBDEs in Chinook salmon. A FEQG of 40 mg·kg^{-1} ww was used to calculate concentrations of ΣHBCDDs in Chinook salmon that are in excess of this Canadian FEQG [40].

It should be noted that the reworked PBDE guideline was based on PBDE homologs found in the Chinook salmon data collected for this present study, which could affect the newly-calculated ΣPBDE FEQG used for contaminant comparisons. As the original FEQG for PBDEs was reported in terms of homologs, however, this was a necessary step in order to obtain a comparative look at the concentrations of ΣPBDEs in Chinook salmon.

The PCB tissue residue guideline was reworked based on the present study's concentrations of PCBs in Chinook salmon. This could also affect the PCB guideline value of 0.0688 mg·kg^{-1} ww. The present study's reworked PCB tissue residue guideline was similar to that calculated by Hickie et al. [20] (0.05 mg/·kg^{-1} ww), which has been widely accepted in the literature as a valid tissue residue guideline for ΣPCBs. The present study's reworked PCB tissue residue guideline can therefore be considered acceptable for making comparisons to concentrations of PCBs [7, 41].

Based on Chinook salmon samples, log-transformed ratios of ΣPCBs divided by the reworked tissue residue guideline (ΣPCBs/guidelines) and ΣPBDEs divided by the newly-calculated ΣPBDEs FEQG (ΣPBDEs/guidelines) were plotted against time (Figure 4.63). A linear regression analysis was also conducted to determine if there was a relationship between the ratio of ΣPCBs/guidelines and time. A linear regression of ΣPBDEs/guidelines in relation to time could not be conducted due to insufficient data (n = 1 in 2002). No significant relationship was found between the ratio of ΣPCBs/guidelines and time (Table 4.26; see Appendix H for individual outcomes).

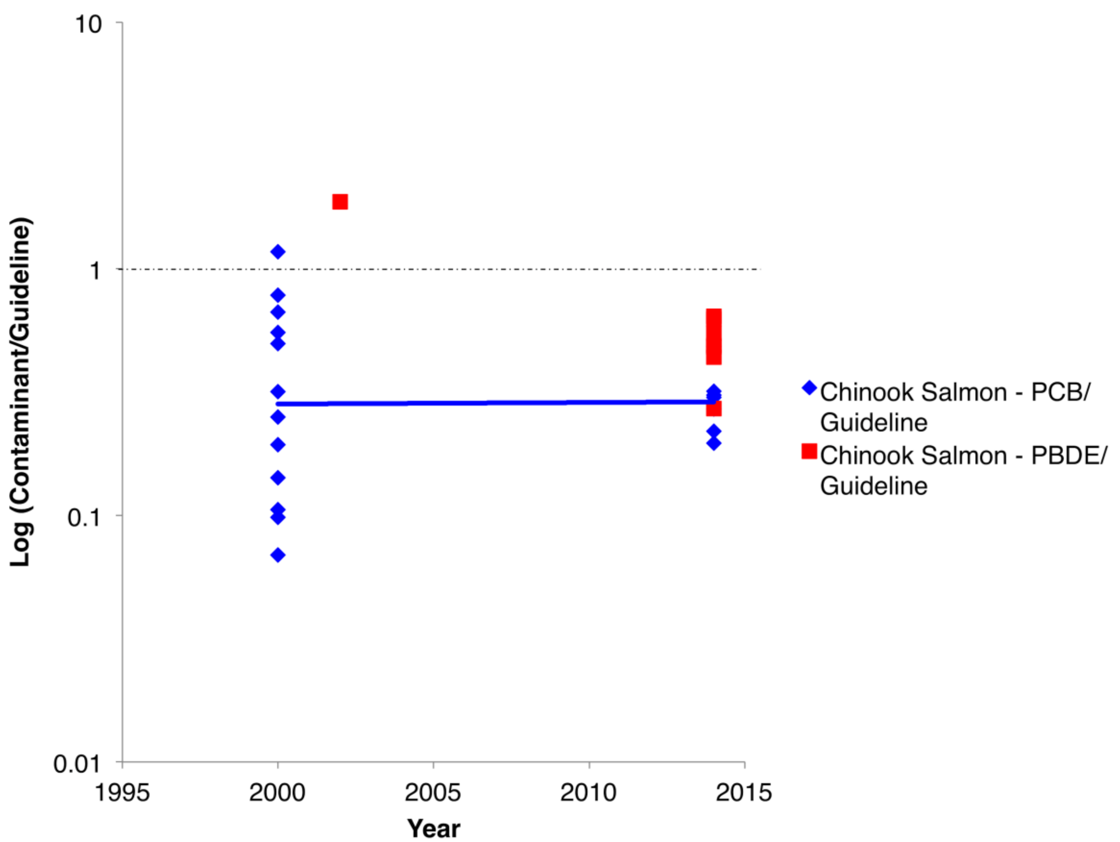

Figure 4.63 Log-transformed ratios of total polychlorinated biphenyls to tissue residue guidelines (ΣPCB/guideline; ◆) and total polybrominated biphenyl ethers to tissue residue guidelines (ΣPBDEs/guideline; ■) in Chinook salmon. The solid line represents the linear regression of the ratio of ΣPCBs/guideline over time. The dashed-dotted line shows when the log-transformed concentrations/guideline are equal to one.

Table 4.26 Results of a linear regression of the ratio of total polychlorinated biphenyls to tissue residue guidelines (ΣPCB/guideline) in Chinook salmon versus time.

Contaminant	Animal	Linear Regression	r^2	P-value
ΣPCBs	Chinook Salmon	Log (ΣPCB/Guideline) = 14.8 - 0.0072*Year	0.0305	0.475

Concentrations of PCBs, PBDEs, and HBCDD in Chinook salmon were divided by their corresponding guidelines in individual Chinook salmon samples collected in 2014 (Figure 4.64; see Appendix H for individual outcomes). Log-transformed geometric mean concentrations of contaminants/guidelines in Chinook salmon sampled in 2014 are depicted in Figure 4.65. All values for the geometric mean concentrations of PCBs, PBDEs, and HBCDD were in excess of their corresponding guidelines (Table 4.27).

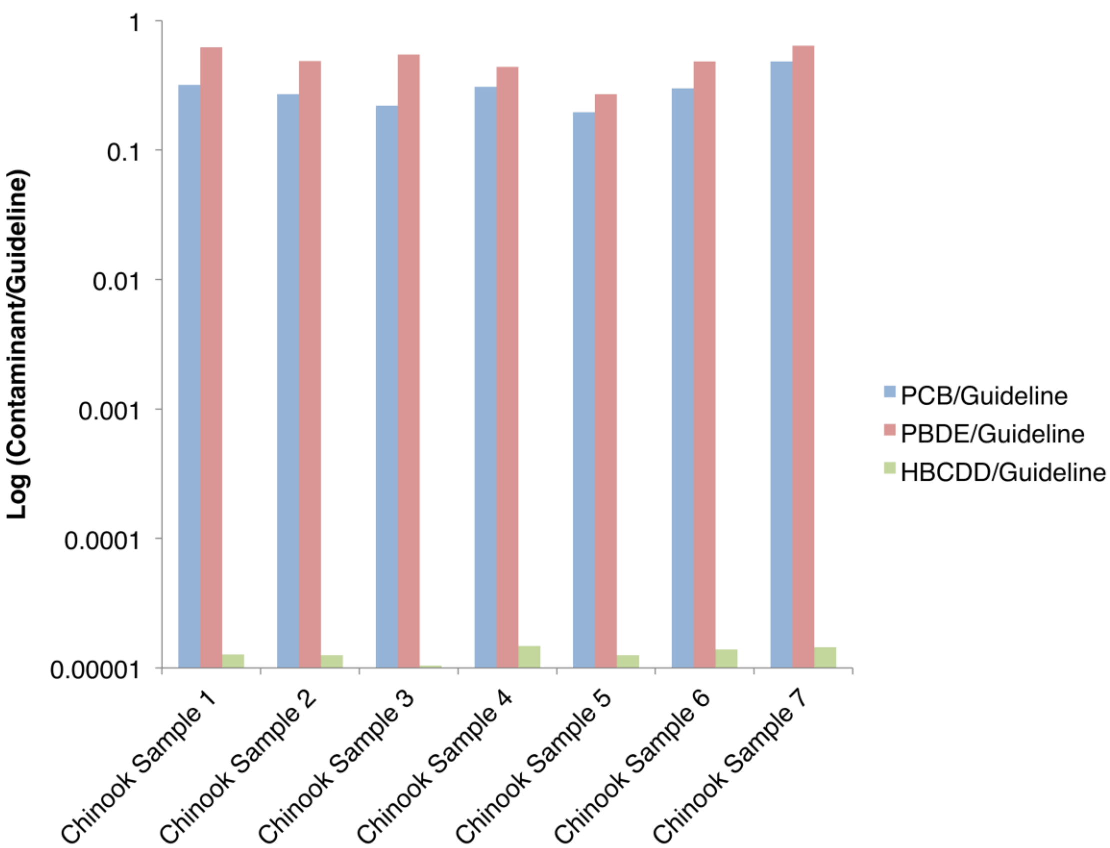

Figure 4.64 The log-transformed ratios of concentrations of polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) in relation to their corresponding guidelines for each Chinook salmon sampled in 2014.

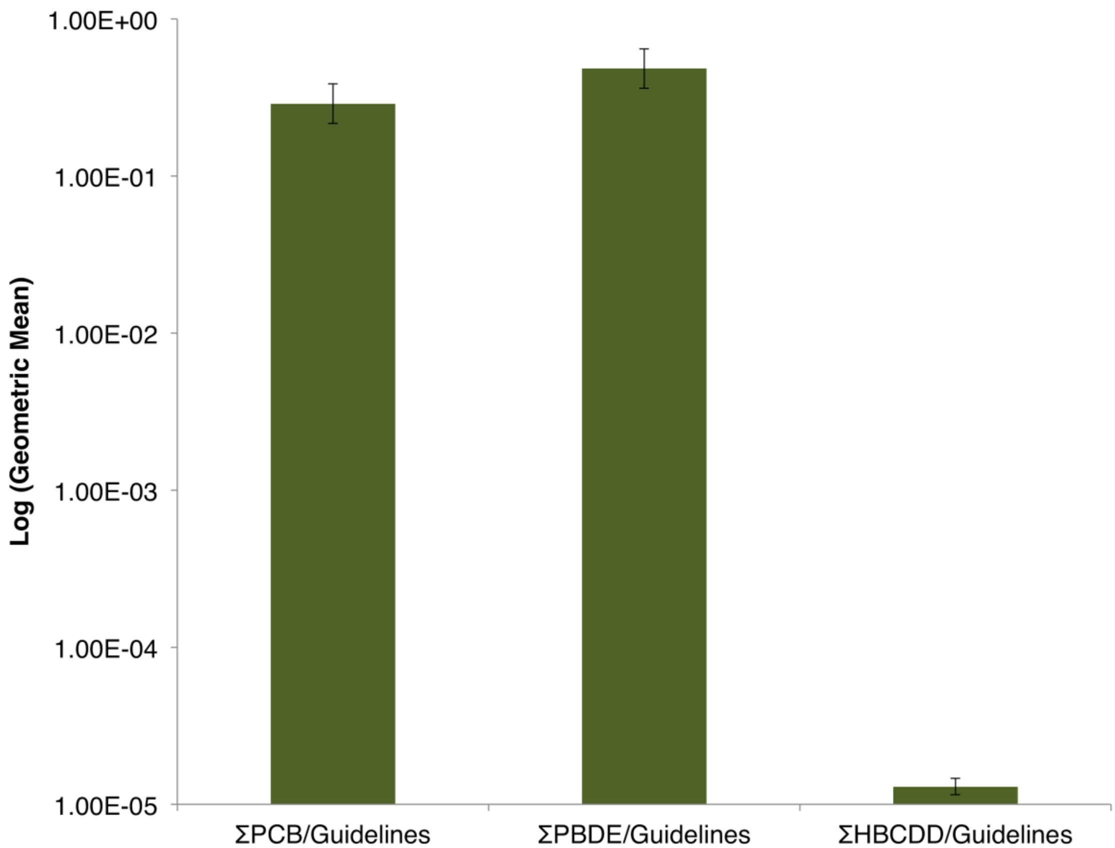

Figure 4.65 The log-transformed geometric mean concentrations of total polychlorinated biphenyls (ΣPCB), total polybrominated diphenyl ethers (ΣPBDE) and total isomers of hexabromocyclododecane (ΣHBCDD) divided by their corresponding Canadian guidelines for wildlife diet.

Table 4.27 The geometric mean values for the concentrations of contaminants in Chinook salmon sampled in 2014 divided by their corresponding Canadian guidelines for wildlife consumption. Upper and lower standard deviations are given. Contaminants investigated were total polychlorinated biphenyls (ΣPCB), total polybrominated diphenyl ethers (ΣPBDE) and total isomers of hexabromocyclododecane (ΣHBCDD).

Ratios	Geometric Mean	Upper SD	Lower SD
ΣPCB/Guidelines	2.89E-01	9.75E-02	7.29E-02
ΣPBDE/Guidelines	4.83E-01	6.45E-01	3.62E-01
ΣHBCDD/Guidelines	1.30E-05	1.46E-05	1.15E-05

Ratios of PBDEs/guidelines were similar in value to ratios of PCBs/guidelines in Chinook salmon (Figure 4.64). The same was true for geometric means of the ratios

(Figure 4.65). This shows that, according to the reworked FEQG for PBDEs, PBDEs are similar in toxicity to PCBs when consumed by mammalian wildlife. The concentrations of HBCDD/guidelines were lower in comparison to the PCBs/guidelines and PBDEs/guidelines, indicating that HBCDD isomers are not as toxic as PCBs and PBDEs.

The finding that PBDEs and PCBs have similar toxicities does not agree with past studies. For example, Hallgren et al.[72] investigated the toxicological effects of PCBs and PBDEs on the thyroid hormones of Sprague-Dawley rats, using Aroclor 1254 and the PBDE congener BDE-47, and found that Aroclor 1254 consistently exhibited more toxic effects than did BDE-47. The discrepancy between that study and the present study could be due to the guidelines used for wildlife consumption, highlighting the possible inaccuracy of these wildlife consumption guidelines for PCBs and PBDEs, which should be noted.

4.3. Risk Evaluation

4.3.1. Cumulative Distributions

Killer Whales

The cumulative distribution of log-transformed ΣPCBs is shown in Figure 4.66, with the average marine mammal TRV for PCBs (1.5 $mg \cdot kg^{-1}$ lw) plotted as a vertical line. Overall, 96.6% of all the concentrations of ΣPCBs measured in killer whales over time were above the average marine mammal TRV for PCBs. This indicates that the majority of killer whales sampled are possibly experiencing health risks from PCBs.

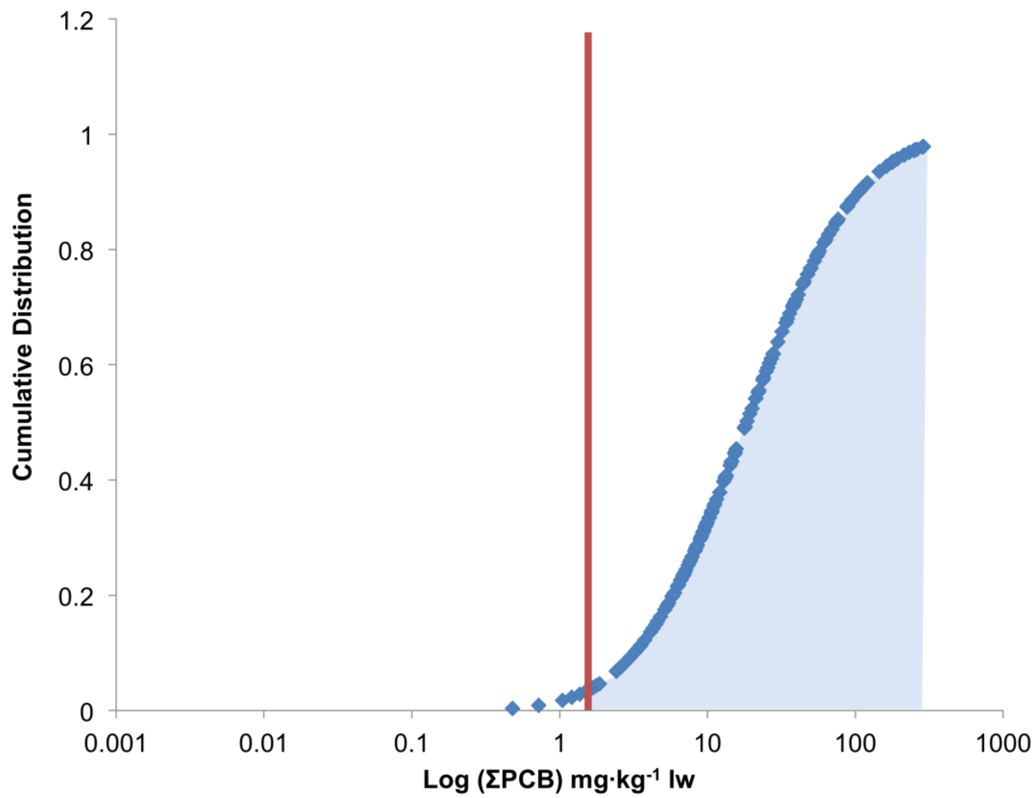

Figure 4.66 Cumulative distribution of the log-transformed concentrations of total polychlorinated biphenyls (ΣPCBs; mg·kg^{-1} lw) in killer whales in relation to the average ΣPCB toxicity reference value (TRV) represented by the vertical line (1.5 mg·kg^{-1} lw).

The cumulative distribution of the log-transformed concentrations of ΣPBDEs (mg·kg^{-1} lw) is presented in Figure 4.67. After comparing the concentrations of PBDEs measured in killer whales to the TRV for ΣPBDEs (1.5 mg·kg^{-1} lw), it was determined that 34.4% of killer whales exhibited concentrations of ΣPBDEs that were above the TRV. This indicates that over a third of the killer whales sampled in this study may be experiencing health risks caused by PBDE contaminants.

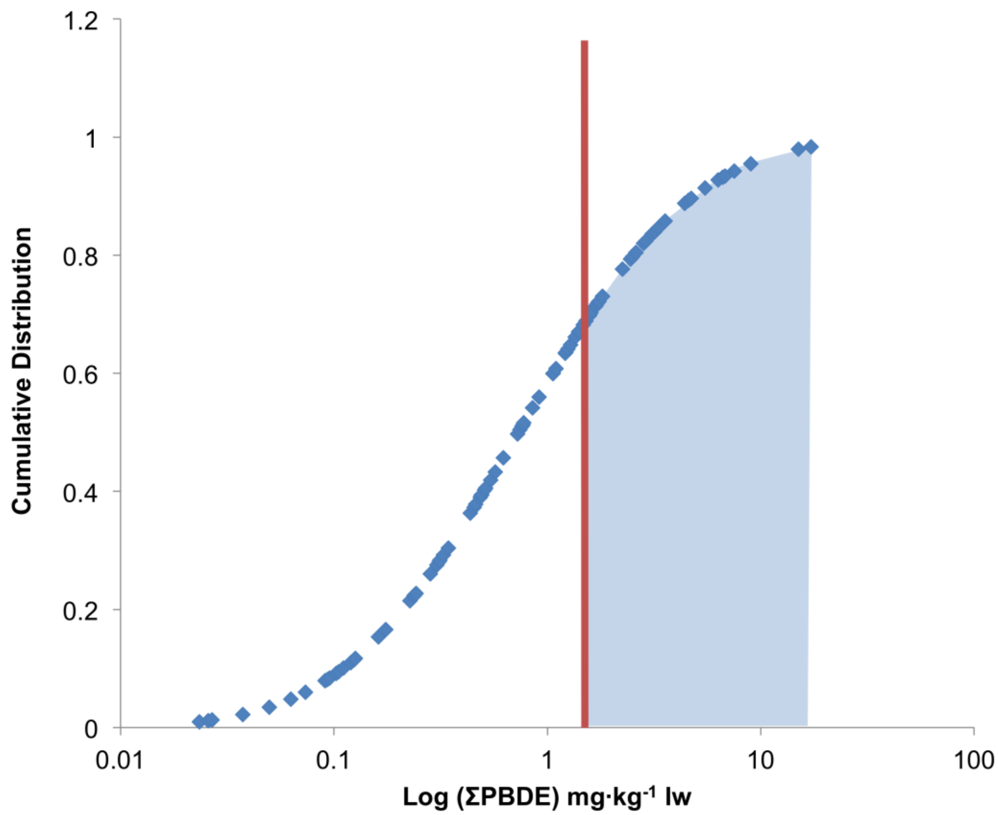

Figure 4.67 Cumulative distribution of the log-transformed concentrations of total polybrominated diphenyl ethers (ΣPBDEs; mg·kg^{-1} lw) in killer whales in relation to the ΣPBDE toxicity reference value (TRV) represented by the vertical line (1.5 mg·kg^{-1} lw).

The cumulative distribution of the log-transformed concentrations of ΣHBCDDs (mg·kg^{-1} lw) is presented in Figure 4.68. The concentrations of ΣHBCDDs measured in killer whales in 2015 were compared to the IC_{50} value for ΣHBCDDs (in mg·kg^{-1} lw). The IC_{50} value for ΣHBCDDs (4 μM) was converted into 1.0 x 10^{-6} mg·kg^{-1} lw. All the concentrations of ΣHBCDDs measured in killer whales were below this IC_{50} value for ΣHBCDDs, indicating that killer whales in the Salish Sea are possibly not experiencing health risks due to the effects of ΣHBCDDs.

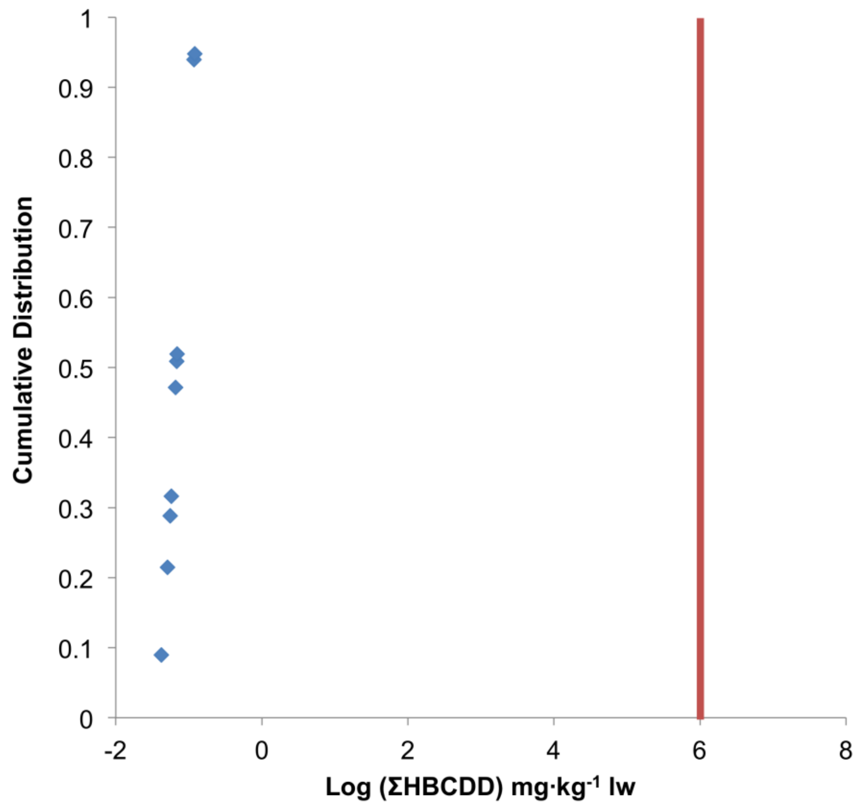

Figure 4.68 Cumulative distribution of the log-transformed concentrations of total isomers of hexabromocyclododecane (ΣHBCDDs; mg·kg^{-1} lw) in killer whales in relation to the 50% inhibition concentration (IC_{50}) value for ΣHBCDDs, represented by the vertical line (1.0 x 10^{-6} mg·kg^{-1} lw).

It should be noted that the aqueous solubility of ΣHBCDDs is 3.4 x 10^{-3} mg·L^{-1}, and thus the IC_{50} value of 4 μM for ΣHBCDDs, which is equivalent to 2.567 mg·L^{-1} (see Methods for calculations), exceeds the aqueous solubility of ΣHBCDDs. A concentration of this magnitude for ΣHBCDDs is therefore not likely to occur in a natural aqueous environment. This is possibly why the observed concentrations of ΣHBCDDs in killer whales are far below the IC_{50} value for ΣHBCDDs. The IC_{50} value of 4 μM was the only reference for the effects of ΣHBCDDs that was available at the time of this study and thus a comparison to the observed concentration of ΣHBCDDs in killer whales was conducted.

ΣPCBs were the group of contaminants that were present in the highest percentage of killer whales at levels that were above marine mammal TRVs. ΣPBDEs had the second highest percentage of killer whales with contaminant concentrations above the marine mammal TRVs. ΣPCBs are therefore possibly causing the most health risks to Salish Sea killer whales.

Chinook Salmon

The cumulative distribution of the log-transformed concentrations of ΣPCBs in Chinook salmon is presented in Figure 4.69. The reworked Canadian ΣPCB tissue residue guideline of 0.0688 mg·kg^{-1} ww[37] was divided by the 14.1% lipid content of Chinook salmon[7] to give 0.488 mg·kg^{-1} lw, and this new ΣPCB tissue residue guideline was log-transformed and plotted as a vertical line in Figure 4.69. All concentrations of ΣPCBs in Chinook salmon over time from this data set were used, and 31.6% of the salmon samples exhibited concentrations of ΣPCBs that were above the reworked Canadian tissue residue guideline for ΣPCBs.

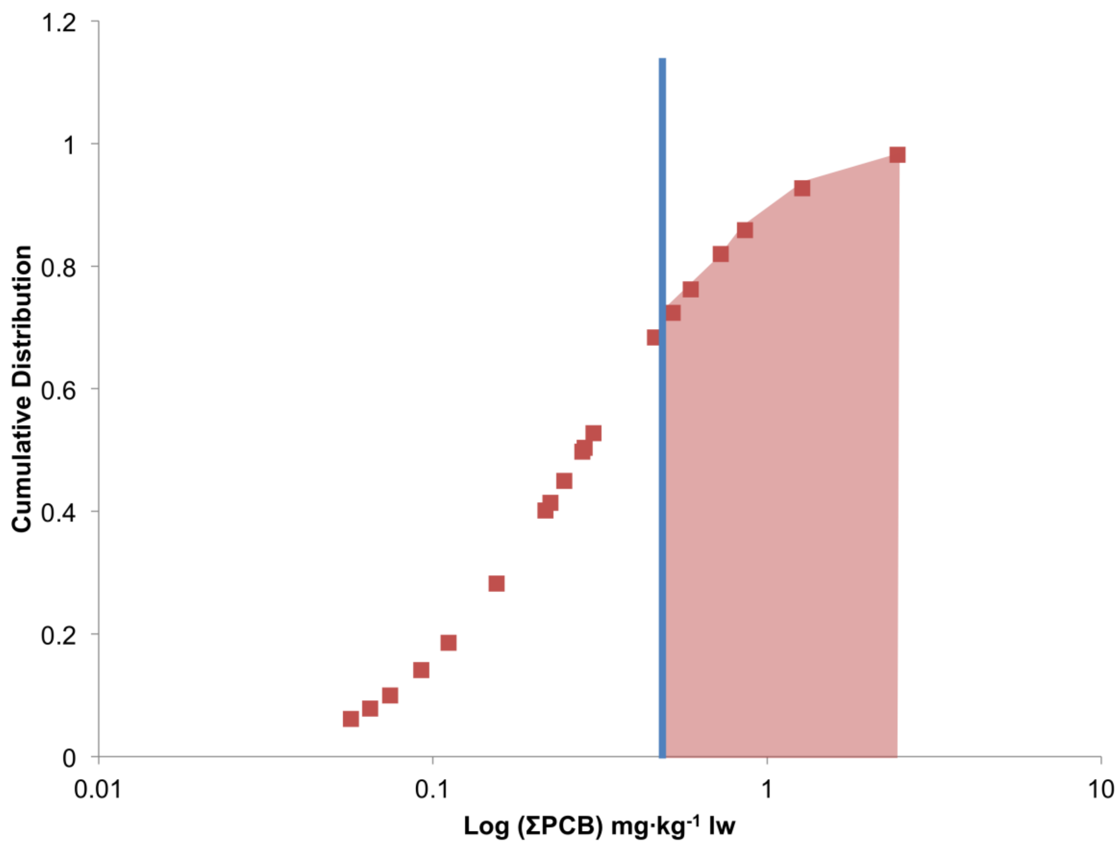

Figure 4.69 Cumulative distribution of the log-transformed concentrations of total polychlorinated biphenyls (ΣPCBs; mg·kg^{-1} lw) in Chinook

salmon in relation to the reworked Canadian ΣPCB tissue residue guideline (0.488 mg·kg^{-1} lw).

The cumulative distribution of the log-transformed concentrations of ΣPBDEs measured in Chinook salmon over time is shown in Figure 4.70. The reworked FEQG for ΣPBDEs [39], calculated to be 0.00945 mg·kg^{-1} ww, was also converted to lipid weight by dividing it by the 14.1% lipid content in Chinook salmon, giving a value of 0.0670 mg·kg^{-1} lw. This new value was plotted as a vertical line in Figure 4.70, and 50.0% of the Chinook salmon that were sampled exhibited concentrations of ΣPBDEs that were above this new FEQG.

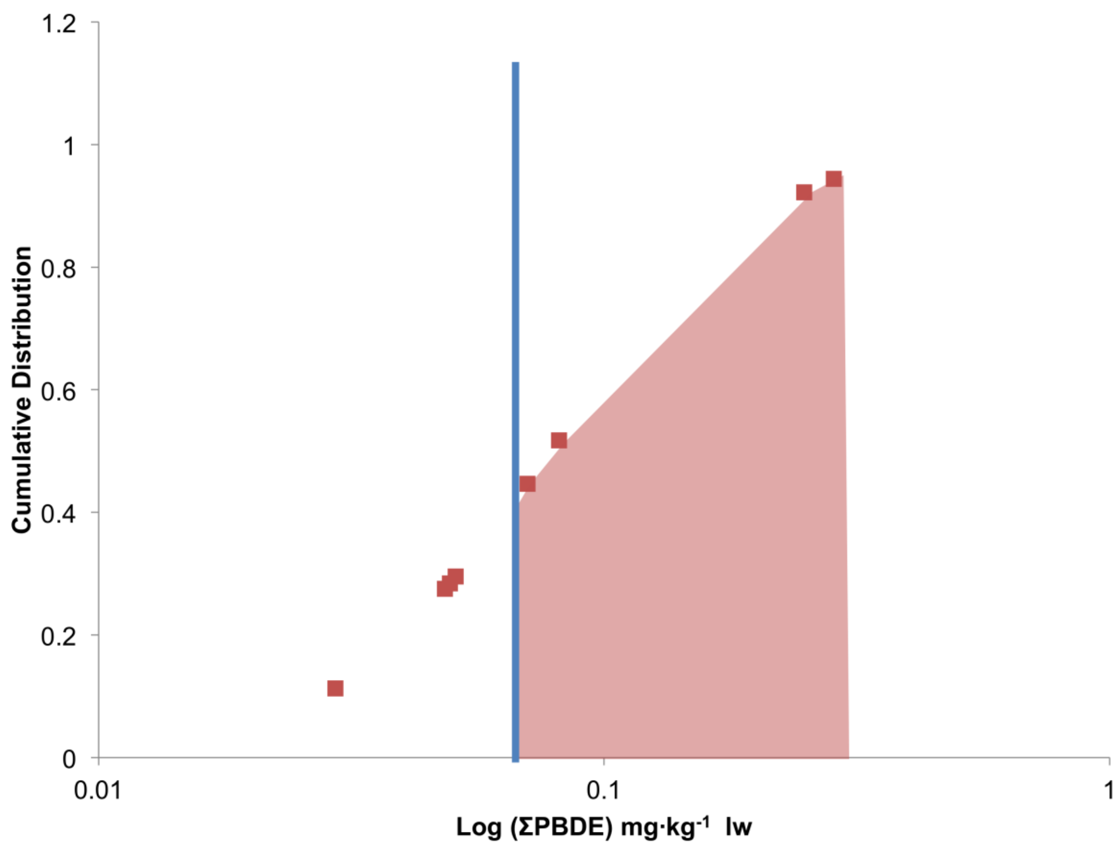

Figure 4.70 Cumulative distribution of the log-transformed concentrations of total polybrominated diphenyl ethers (ΣPBDEs; mg·kg^{-1} lw) in Chinook salmon in relation to the newly-calculated federal environmental quality guideline for ΣPBDEs for fish-eating wildlife (0.0670 mg·kg^{-1} lw).

The cumulative distribution of log-transformed concentrations of ΣHBCDDs in Chinook salmon is depicted in Figure 4.71. The FEQG for ΣHBCDDs (40.0 mg·kg^{-1} ww

[40]) was converted to 2.84 x 10^2 mg·kg^{-1} lw through division by the 14.1% lipid content of Chinook salmon, and this new FEQG value was plotted as a vertical line in Figure 4.71. None of the concentrations of ΣHBCDDs measured in Chinook salmon were found to be above the FEQG for ΣHBCDDs.

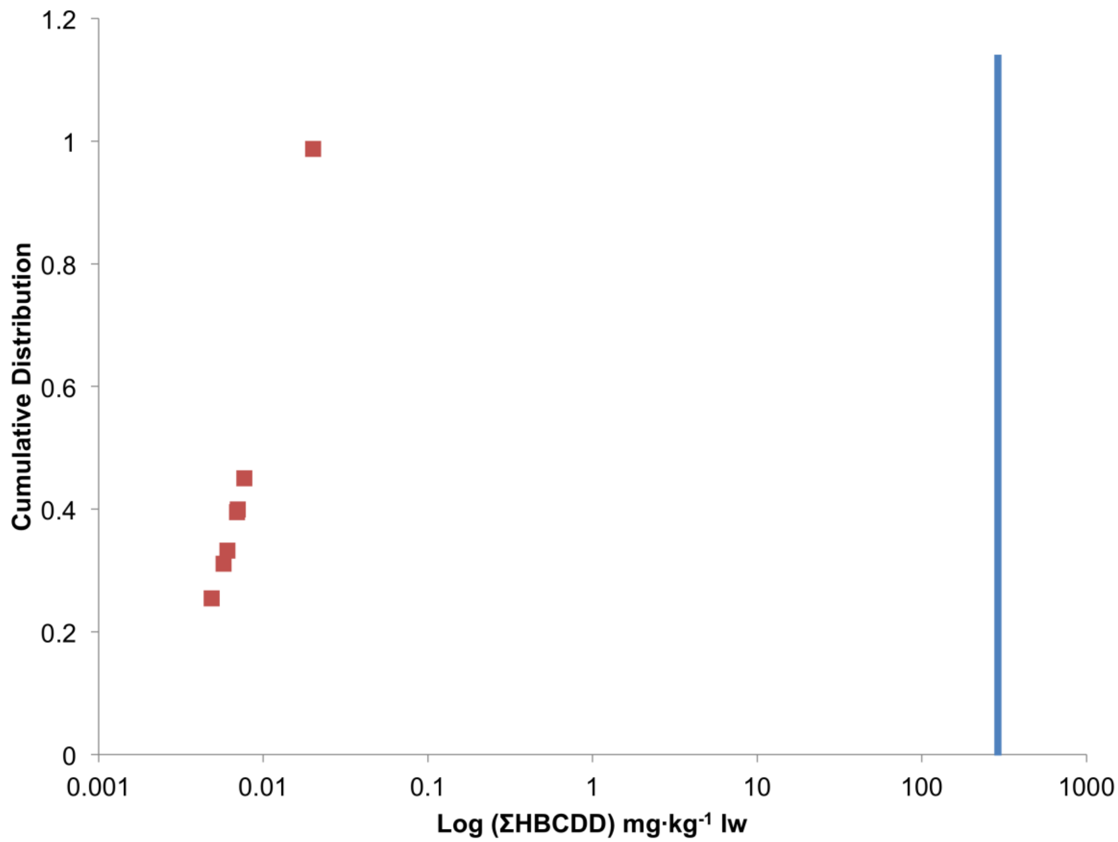

Figure 4.71 Cumulative distribution of the log-transformed concentrations of total hexabromocyclododecane isomers (ΣHBCDDs; mg·kg^{-1} lw) in Chinook salmon in relation to the federal environmental quality guideline for ΣHBCDDs for fish-eating wildlife (2.84 x 10^2 mg·kg^{-1} lw).

A higher percentage of samples of Chinook salmon were found to contain levels of ΣPBDEs that could be considered toxic for fish-eating wildlife than were found to contain toxic levels of ΣPCBs. This does not match the percentage of killer whales that were found to exhibit contaminant levels above the TRVs. This could be because the FEQG for ΣPBDEs for fish-eating wildlife was more conservative than the tissue residue guideline for ΣPCBs. Furthermore, this finding does not agree with other studies, such as that of Hallgren et al. [16]. Further investigation should be conducted to better understand the accuracy of the FEQG for ΣPBDEs for fish-eating wildlife.

4.3.2. BMFs

Biomagnification factors (BMFs) for ΣPCBs, ΣPBDEs, and ΣHBCDDs were calculated separately for each sex and ecotype of Salish Sea killer whales. The calculated BMFs were based on measured concentrations of PCBs in resident killer whales and their predominant prey, Chinook salmon, and were compared to modelled BMFs of ΣPCBs, calculated previously by Alava et al. [7]. The calculated log-transformed BMFs of ΣPCBs for male and female northern resident killer whales were determined to be 1.54±0.563 and 1.39±0.662, respectively, whereas the log-transformed BMFs of ΣPCBs for male and female southern resident killer whales were 1.77±0.595 and 1.81±0.668, respectively (Figure 4.72). The predicted log-transformed BMFs of ΣPCBs, taken from Alava et al. [7], were 2.23±0.330 and 1.54±0.330, respectively, for male and female northern resident killer whales, and 2.22±0.330 and 2.15±0.330, respectively, for male and female southern resident killer whales. All modelled and observed log-transformed BMFs of ΣPCBs for male and female northern and southern resident killer whales are presented in Table 4.28, along with corresponding standard deviations.

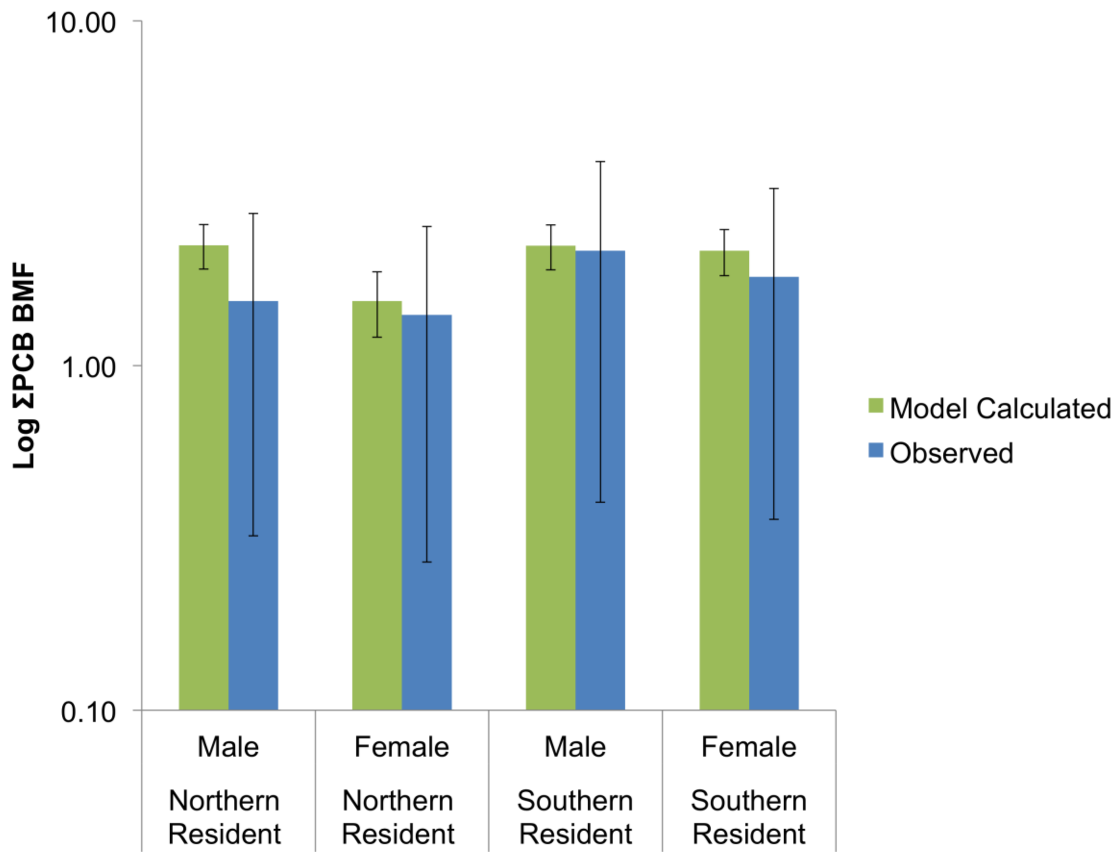

Figure 4.72 **Log-transformed biomagnification factors (BMF) for concentrations of total polychlorinated biphenyls (ΣPCBs) found in male and female northern and southern resident killer whales, in comparison to the modelled BMFs of ΣPCBs for those whales, based on the model from Alava et al.[7]. Error bars represent the standard deviation.**

Table 4.28 **Predicted and observed log-transformed biomagnification factors (BMF) for total polychlorinated biphenyls (ΣPCBs) in male and female northern (NR) and southern (SR) resident killer whales. Corresponding log-transformed standard deviations are included.**

Sex	Ecotype	Contaminant	Type of Data	Log ΣPCB BMF	Log Standard Deviation (±)
Male	NR	PCBs	Predicted	2.23E+00	3.30E-01
Female	NR	PCBs	Predicted	1.54E+00	3.30E-01
Male	SR	PCBs	Predicted	2.22E+00	3.30E-01
Female	SR	PCBs	Predicted	2.15E+00	3.30E-01
Male	NR	PCBs	Observed	1.54E+00	1.22E+00
Female	NR	PCBs	Observed	1.40E+00	1.13E+00
Male	SR	PCBs	Observed	2.15E+00	1.75E+00

Sex	Ecotype	Contaminant	Type of Data	Log ΣPCB BMF	Log Standard Deviation (±)
Female	SR	PCBs	Observed	1.81E+00	1.45E+00

The log-transformed BMF values of ΣPBDEs were calculated to be 0.438±0.718 and 0.599±0.595 for female and male northern resident killer whales, respectively (Figure 4.73), and to be 1.33±0.549 and 1.62±0.451 for male and female southern resident killer whales, respectively (Figure 4.74). Modelled BMF values of ΣPBDEs in this study were not calculated following Alava et al. [64]'s ΣPBDE bioaccumulation model for killer whales, but rather through the use of the ΣPBDE bioaccumulation model outputs from Alava et al. [64]. The bioaccumulation model from Alava et al. [64] gave predicted concentrations of ΣPBDEs only for male and female resident killer whales as a group, not specifically for northern or southern ecotypes; therefore, predicted log-transformed BMF values of ΣPBDEs were only calculated for male and female resident killer whales as a single group. The modelled log-transformed BMFs for ΣPBDEs, based on the study by Alava et al. [64], were calculated to be 0.610±0.669 and 0.539±0.632 for male and female resident killer whales, respectively. The observed and modelled log-transformed BMF values of ΣPBDEs for resident killer whales are outlined in Table 4.29, along with their corresponding log-transformed standard deviations. A short-coming of these BMF calculations for ΣPBDEs was the large standard deviation ranges, which indicated that there was some uncertainty involved and conclusions deduced from these ΣPBDE BMFs should be considered with caution.

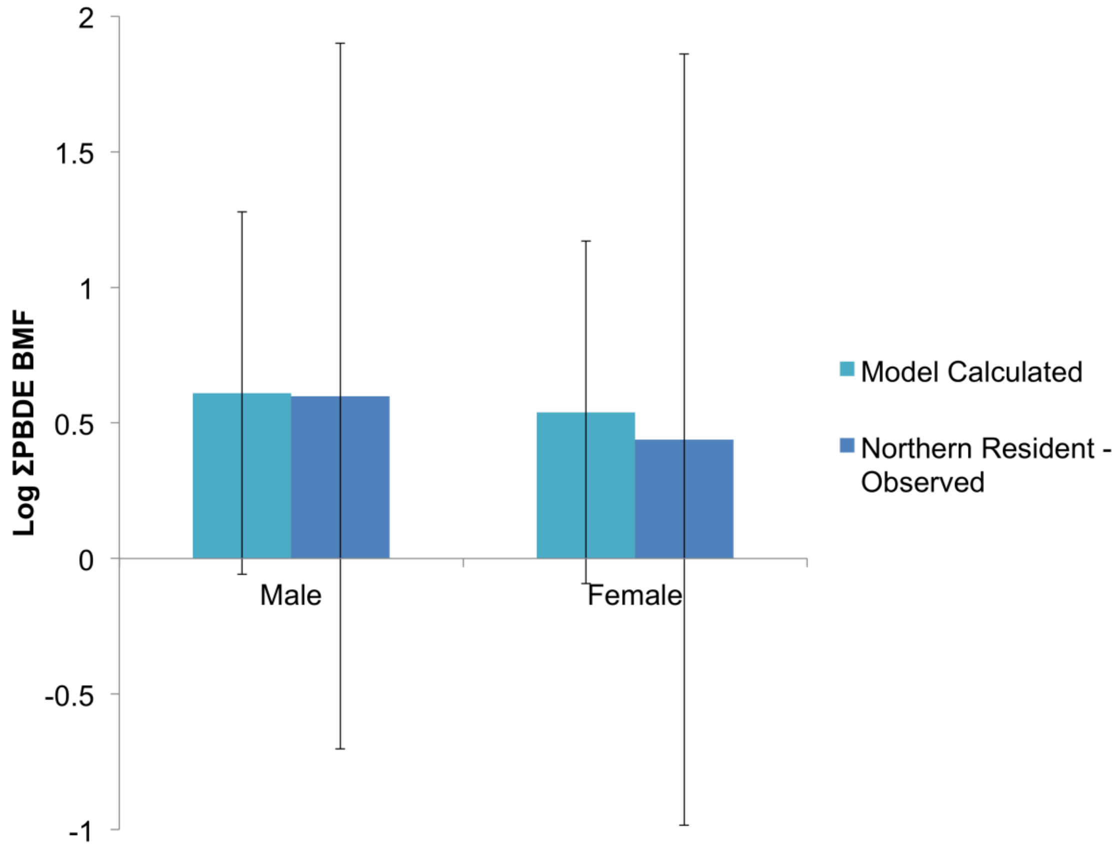

Figure 4.73 **Log-transformed biomagnification factors (BMF) for concentrations of total polybrominated diphenyl ethers (ΣPBDEs) found in male and female northern resident killer whales, in comparison to modelled BMFs of ΣPBDEs for male and female resident killer whales, based on the model from Alava et al.[64]. Error bars represent the standard deviation.**

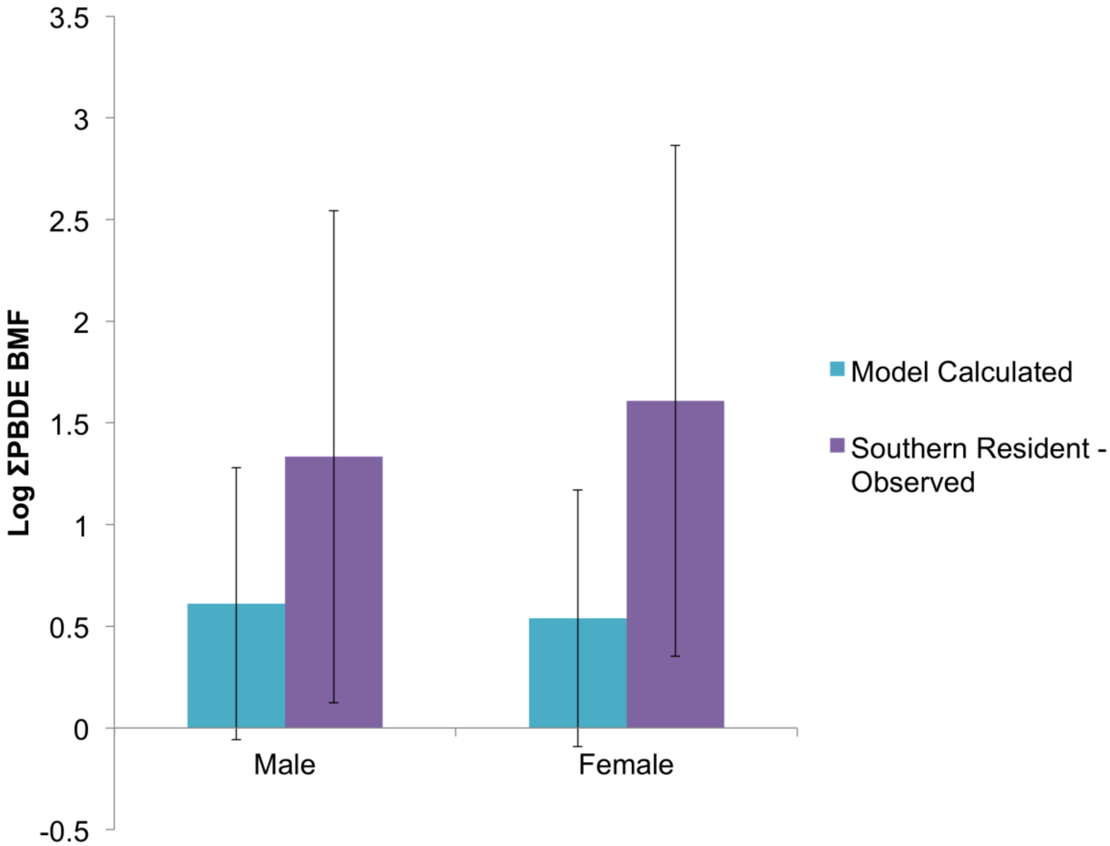

Figure 4.74 **Log-transformed biomagnification factors (BMF) for concentrations of total polybrominated diphenyl ethers (ΣPBDEs) found in male and female southern resident killer whales, in comparison to modelled BMFs of ΣPBDEs for resident male and female resident killer whales, based on the model from Alava et al. [64]. Error bars represent the standard deviation.**

Table 4.29 **Predicted and observed log-transformed biomagnification factors (BMF) for total polybrominated diphenyl ethers (ΣPBDEs) in male and female northern (NR) and southern (SR) resident killer whales. Predicted BMFs treat NR and SR killer whales as a single ecotype. Corresponding log-transformed standard deviations are included.**

Sex	Ecotype	Contaminant	Type of Data	Log ΣPBDE BMF	Log Standard Deviation (±)
Male	Resident	PBDEs	Predicted	6.10E-01	6.69E-01
Female	Resident	PBDEs	Predicted	5.39E-01	6.32E-01
Male	NR	PBDEs	Observed	5.99E-01	1.30E+00
Female	NR	PBDEs	Observed	4.38E-01	1.42E+00
Male	SR	PBDEs	Observed	1.33E+00	1.21E+00

Sex	Ecotype	Contaminant	Type of Data	Log ΣPBDE BMF	Log Standard Deviation (±)
Female	SR	PBDEs	Observed	1.61E+00	1.26E+00

The log-transformed BMF values for ΣHBCDDs were calculated to be 0.675±0.192 for male southern resident killer whales and 0.816±0.246 for females (Figure 4.75). No comparison was made to modelled BMF values for ΣHBCDDs because there were no modelled values available at the time of this study. The observed log-transformed BMF values for ΣHBCDDs for southern resident killer whales and their corresponding log-transformed standard deviations are presented in Table 4.30.

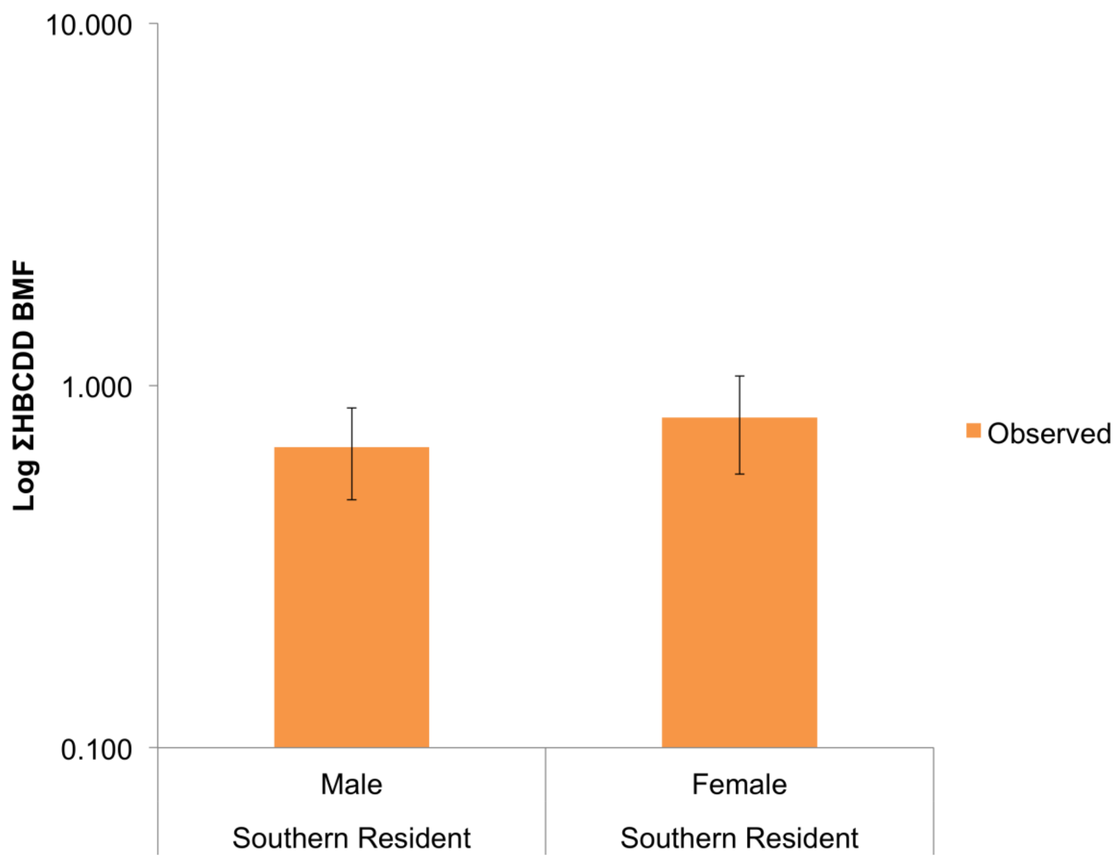

Figure 4.75 Observed log-transformed biomagnification factors (BMF) of total isomers of hexabromocyclododecane (ΣHBCDD) for male and female southern resident killer whales. Error bars represent the standard deviation.

Table 4.30 Log-transformed biomagnification factors (BMFs) of total isomers of hexabromocyclododecane (ΣHBCDD) for male and female southern resident killer whales. Corresponding log-transformed standard deviations are included.

Sex	Ecotype	Contaminant	Type of Data	Log ΣHBCDD BMFs	Log Standard Deviation (±)
Male	Southern Resident	ΣHBCDD	Observed	6.75E-01	1.92E-01
Female	Southern Resident	ΣHBCDD	Observed	8.16E-01	2.46E-01

The BMF values for this study were calculated in order to have relevant BMFs based on observed data for resident killer whales. However, it should be noted that when exploring contaminants such as ΣPCBs, ΣPBDEs and ΣHBCDDs, which all are compiled from multiple congeners, the congener profiles of these contaminants might vary among resident killer whales and Chinook salmon. There may be uncertainty in the BMF calculations due to the differing congener profiles for killer whales and Chinook salmon. This discrepancy in congener profiles however is not a main concern when exploring the contaminant ratios from Figures 4.54 – 4.59, especially the ratio profiles exploring contaminants in Chinook salmon and their respective guidelines. This is because the guidelines were calculated using Chinook salmon data. Therefore the congener profiles used for the ratios are the same. The observed BMFs however, were in good accordance with the modelled BMFs calculated based on the work of Alava et al. [7, 64], showing that the BMFs of ΣPCBs, ΣPBDEs, and ΣHBCDDs for resident killer whales are good predictors for understanding the concentrations of those contaminants in resident killer whales, based on concentrations of the same contaminants in Chinook salmon.

4.3.3. Daily Exposure Rates, Hazard Indices and Carcinogenic Risk Estimates

Daily exposure rates based on killer whale body weight were investigated and the contaminants with the highest daily exposure rates were PCBs (Figures 4.76 – 4.82; Table 4.31). Killer whales faced the highest daily exposure rates for all the contaminants during their juvenile life stage. This was because daily exposure rate was based on the weight of the killer whale, with juveniles weighing 1000 kg.

Hickie et al.[20] noted that to protect 95% of the resident killer whale population, the prey of killer whales (Chinook salmon) would have to contain concentrations of PCBs below 8.0 μg·kg^{-1} ww. This value was based on the 17 mg·kg^{-1} threshold value for harbour seals, the least conservative value of all the PCB toxicity reference values (see Table 1.1). Exposure rates were calculated based on this suggested concentration of 8.0 μg·kg^{-1} ww of PCBs. These rates are reported for each resident killer whale life stage in Table 4.31. All daily exposure rates for PCBs from the present study were far above the exposure rates based on Hickie et al.[20]'s suggested concentration intake of PCBs. This means that 95% of the killer whale population was not protected from serious health impacts, such as effects on the immune, endocrine, and hepatic systems, caused by ingestion of PCBs through the consumption of Chinook salmon (Table 1.1).

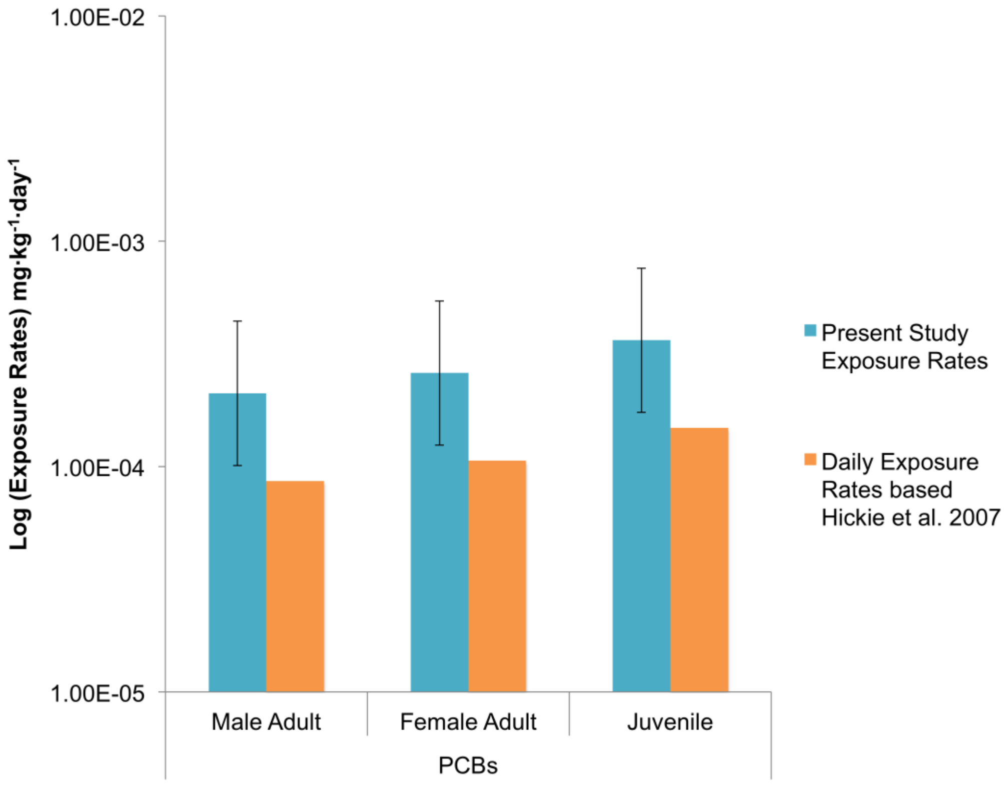

Figure 4.76 Log-transformed daily exposure rates (mg·kg^{-1}·day^{-1}) to polychlorinated biphenyls (PCBs), based on the average weights of resident killer whales in three different life stages. Exposure rates based on the present study's findings of PCB levels in Chinook salmon are shown in comparison to the concentrations of PCBs (8.0 μg·kg^{-1} ww) in Chinook salmon that were suggested by Hickie et al. (2007) to be necessary in order to protect 95% of the resident killer whale population. Error bars represent the standard deviation.

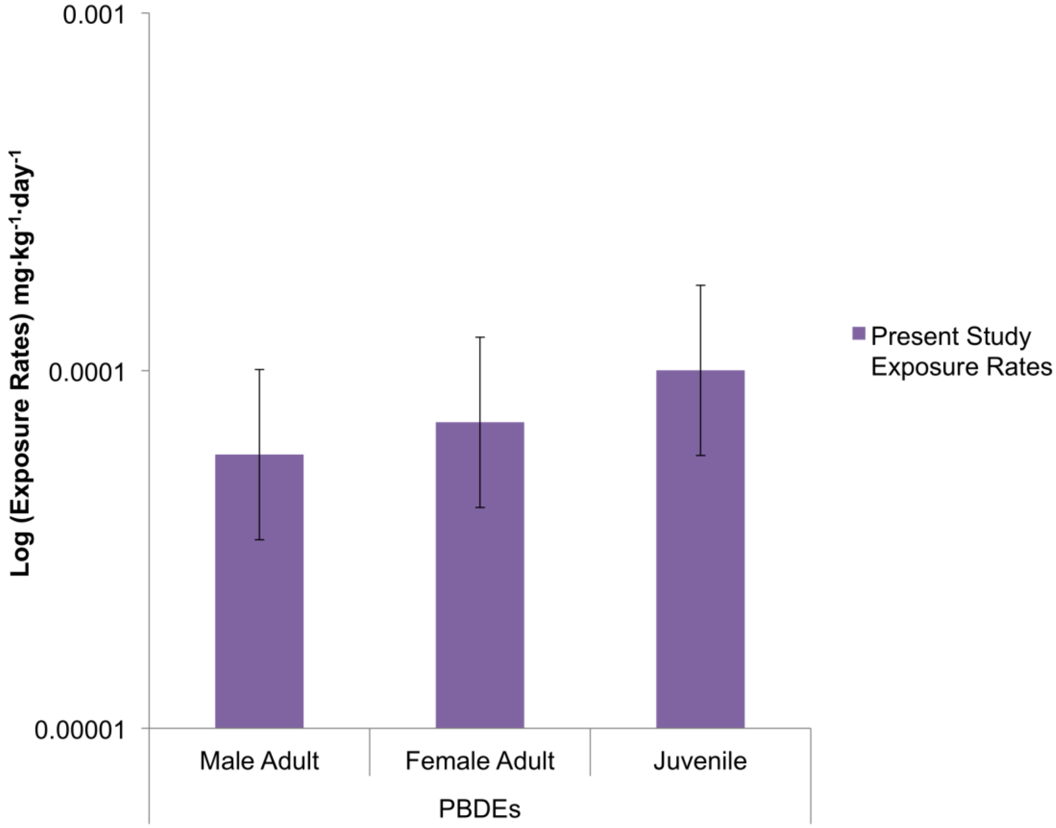

Figure 4.77 Log-transformed daily exposure rates (mg·kg^{-1}·day^{-1}) to polybrominated biphenyl diethers (PBDEs), based on the average weights of resident killer whales in three different life stages. Error bars represent the standard deviation.

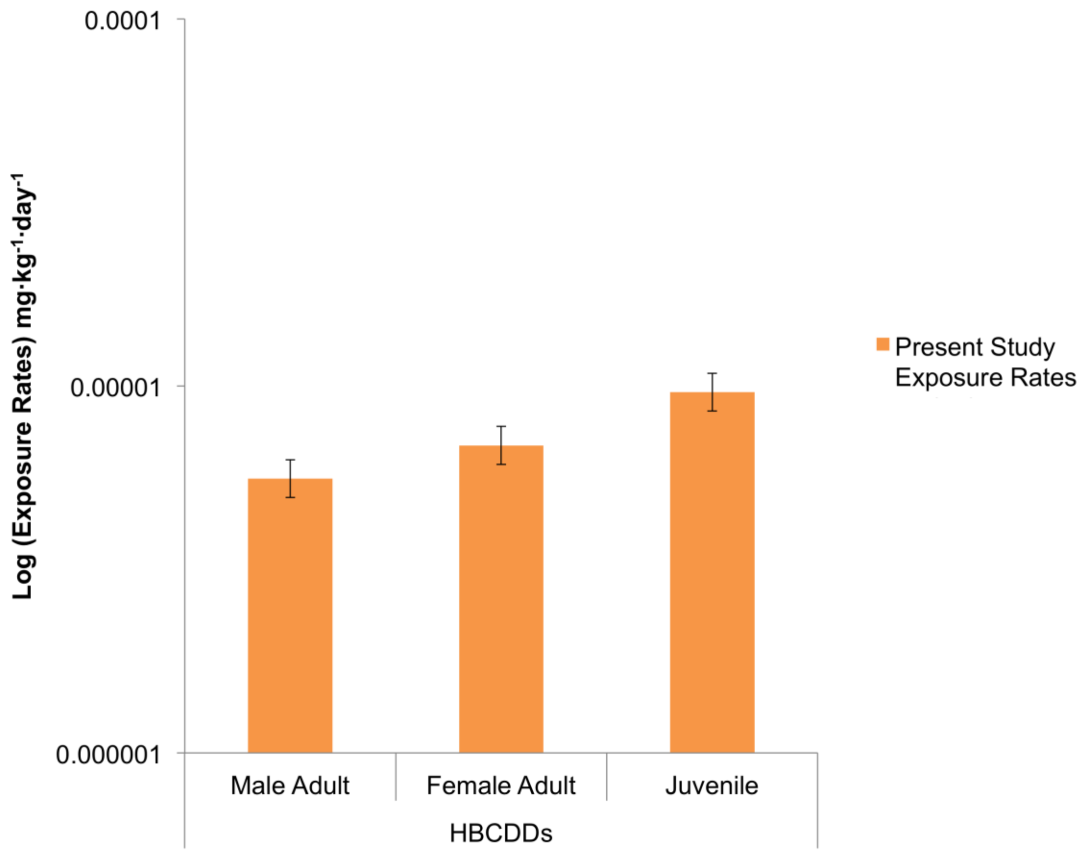

Figure 4.78 Log-transformed daily exposure rates (mg·kg^{-1}·day^{-1}) to hexabromocyclododecane isomers (HBCDDs), based on the average weights of resident killer whales in three different life stages. Error bars represent the standard deviation.

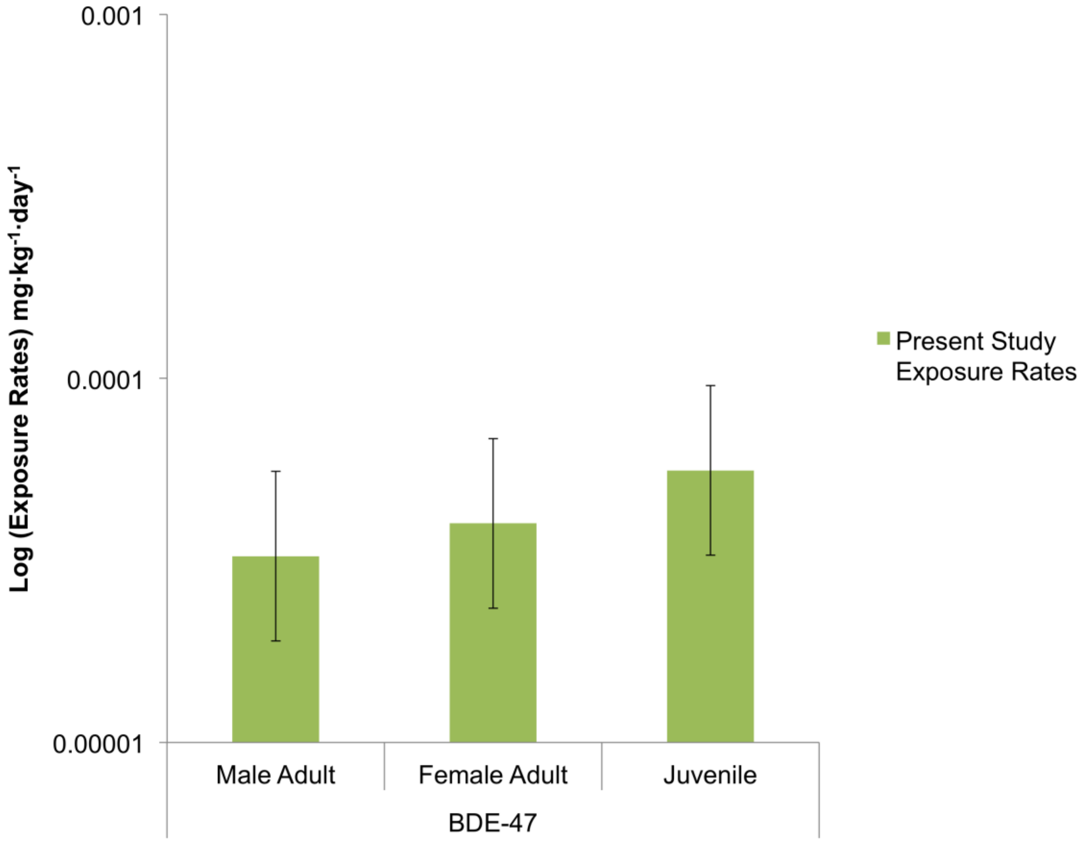

Figure 4.79 **Log-transformed daily exposure rates ($mg \cdot kg^{-1} \cdot day^{-1}$) to the polybrominated diphenyl ether congener BDE-47, based on the average weights of resident killer whales in three different life stages. Error bars represent the standard deviation.**

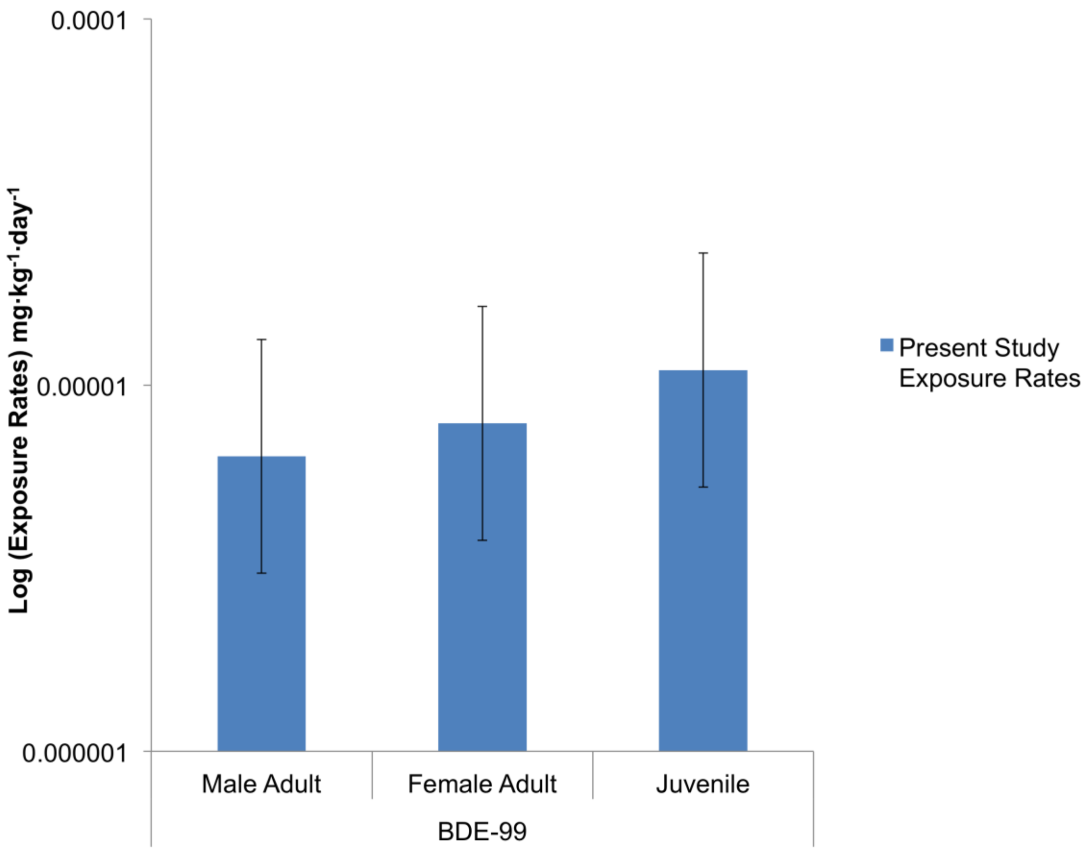

Figure 4.80 **Log-transformed daily exposure rates (mg·kg^{-1}·day^{-1}) to the polybrominated diphenyl ether congener BDE-99, based on the average weights of resident killer whales in three different life stages. Error bars represent the standard deviation.**

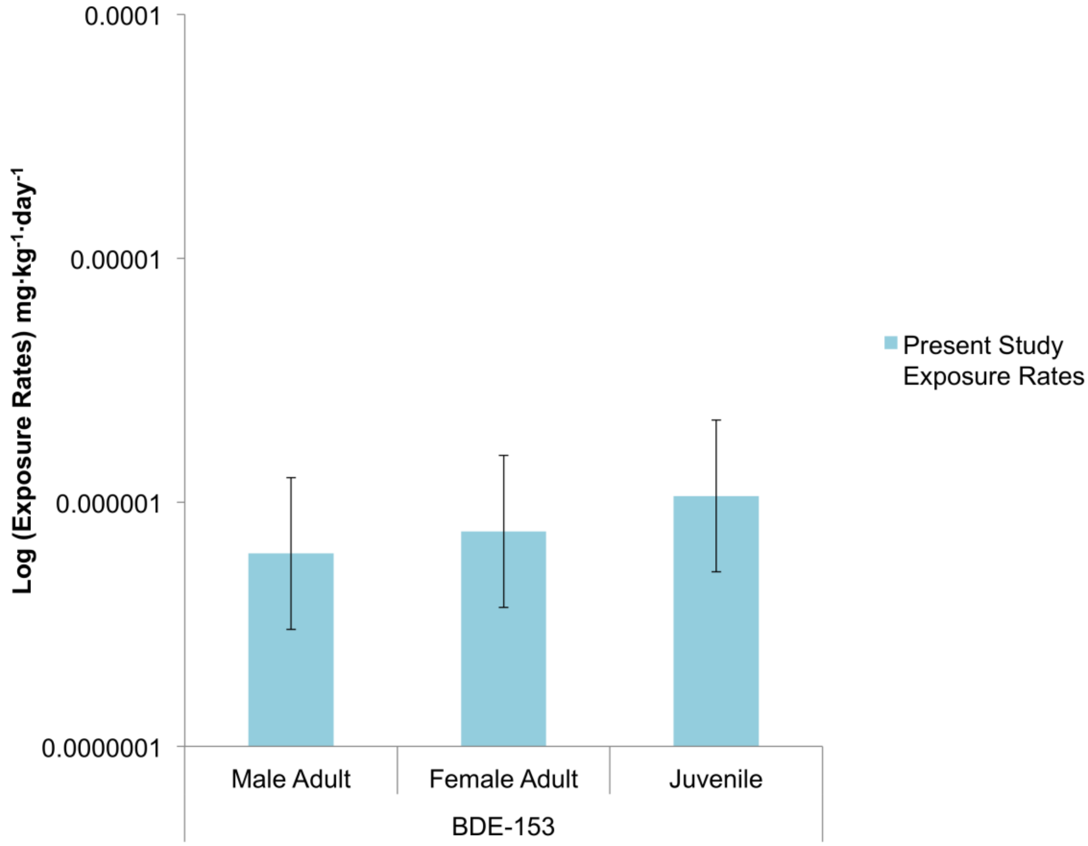

Figure 4.81 **Log-transformed daily exposure rates (mg·kg^{-1}·day^{-1}) to the polybrominated diphenyl ether congener BDE-153, based on the average weights of resident killer whales in three different life stages. Error bars represent the standard deviation.**

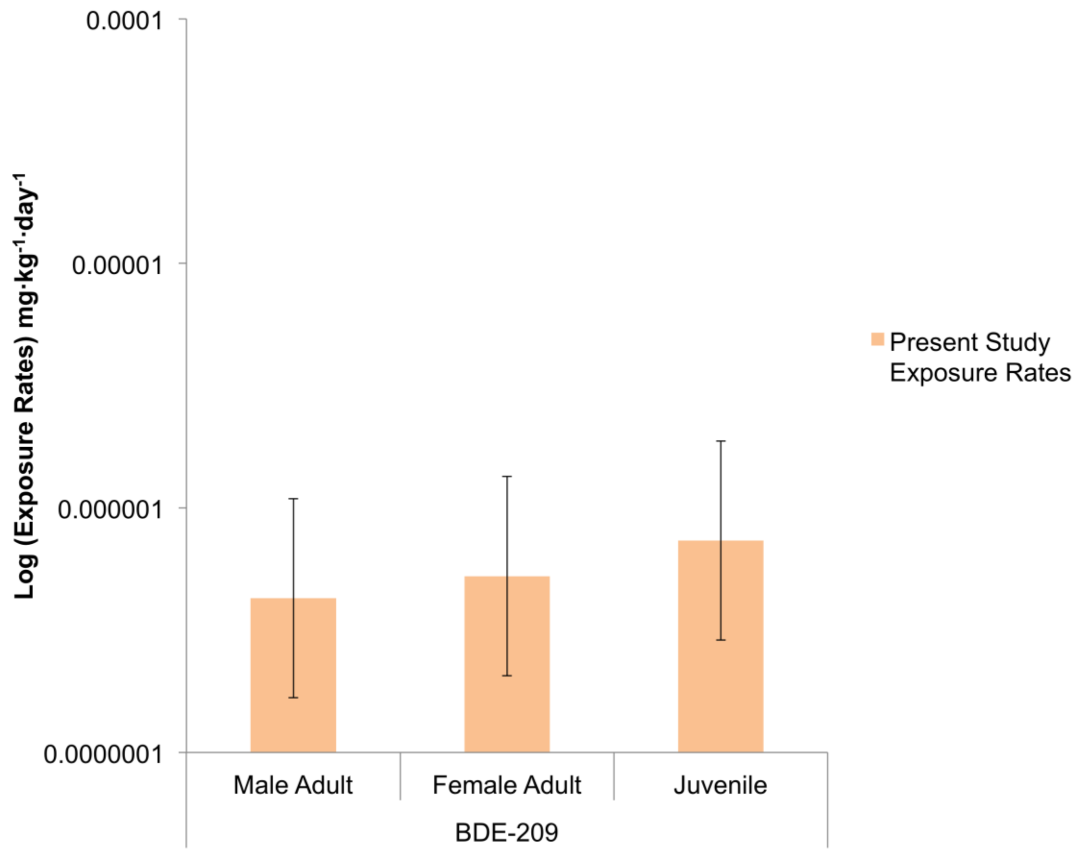

Figure 4.82 **Log-transformed daily exposure rates ($mg \cdot kg^{-1} \cdot day^{-1}$) to the polybrominated diphenyl ether congener BDE-209, based on the average weights of resident killer whales in three different life stages. Error bars represent the standard deviation.**

Table 4.31 **Daily exposure rates to seven contaminants in relation to the sex and weight of resident killer whales. The contaminants investigated were polychlorinated biphenyls (PCBs), hexabromocyclododecane isomers (HBCDDs), polybrominated diphenyl ethers (PBDEs), and the PBDE congeners BDE- 47, BDE-99, BDE 153 and BDE 209.**

Contaminant	Sex & Life Stage	Estimated Daily Exposure Rate ($mg \cdot kg^{-1} \cdot day^{-1}$)	Upper Standard Deviation	Lower Standard Deviation
PCBs	Male Adult	2.11E-04	4.42E-04	1.01E-04
	Female Adult	2.60E-04	5.44E-04	1.24E-04
	Juvenile	3.63E-04	7.60E-04	1.74E-04
PCBs*	Male Adult	8.62E-05	N/A	N/A
	Female Adult	1.06E-04	N/A	N/A

Contaminant	Sex & Life Stage	Estimated Daily Exposure Rate ($mg \cdot kg^{-1} \cdot day^{-1}$)	Upper Standard Deviation	Lower Standard Deviation
	Juvenile	1.48E-04	N/A	N/A
PBDEs	Male Adult	5.83E-05	1.01E-04	3.37E-05
	Female Adult	7.17E-05	1.24E-04	4.14E-05
	Juvenile	1.00E-04	1.73E-04	5.79E-05
HBCDDs	Male Adult	5.59E-06	6.30E-06	4.96E-06
	Female Adult	6.88E-06	7.75E-06	6.11E-06
	Juvenile	9.62E-06	1.08E-05	8.54E-06
BDE-47	Male Adult	3.25E-05	5.56E-05	1.90E-05
	Female Adult	4.00E-05	6.84E-05	2.34E-05
	Juvenile			
		5.59E-05	9.56E-05	3.27E-05
BDE-99	Male Adult	6.39E-06	1.33E-05	3.06E-06
	Female Adult	7.86E-06	1.64E-05	3.77E-06
	Juvenile	1.10E-05	2.29E-05	5.27E-06
BDE-153	Male Adult	6.17E-07	1.26E-06	3.02E-07
	Female Adult	7.60E-07	1.56E-06	3.71E-07
	Juvenile	1.06E-06	2.17E-06	5.19E-07
BDE-209	Male Adult	4.27E-07	1.09E-06	1.67E-07
	Female Adult	5.26E-07	1.34E-06	2.06E-07
	Juvenile	7.35E-07	1.88E-06	2.88E-07

*based on Hickie et al. [20]'s suggested intake concentration of 8.0 $\mu g \cdot kg^{-1}$ ww of PCBs

The hazard index outcomes for various reference doses are depicted in Figures 4.83 – 4.88 and in Table 4.32. Due to there being no reference dose for ΣPCBs, the reference doses for Aroclor 1254 and 1016 were used. Aroclor 1254 and 1016 are different mixtures of ΣPCBs and thus may not be identical to the ΣPCB contaminants that killer whales are experiencing; however, hazard indices for Aroclor 1254 and 1016 were calculated to understand if the levels of ΣPCBs were hazardous compared to the reference doses for Aroclor 1254 and 1016. All hazard indices calculated for the reference doses for Acoclor 1254 and 1016 were above one, indicating that resident killer whales are experiencing daily exposure rates of ΣPCBs that are hazardous to their health at all life stages. Overall, resident killer whales may potentially be experiencing adverse non-carcinogenic affects to their health.

Hazard indices calculated for BDE-47, BDE-99, BDE-153, and BDE-209 were all below one, and therefore these contaminants were considered not to be hazardous to the health of resident killer whales.

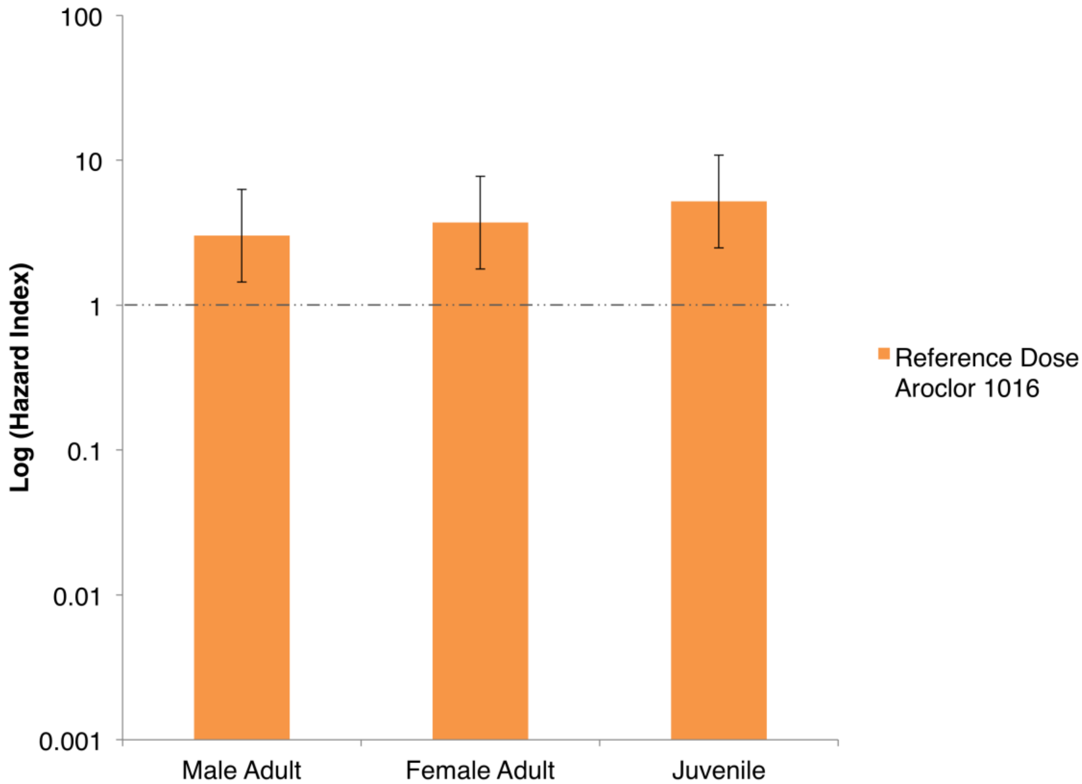

Figure 4.83 Log-transformed hazard indices for polychlorinated biphenyls (PCBs) in three different life stages of resident killer whales, based on the reference dose for Aroclor 1016. The dashed-dotted line shows when the hazard index is equal to one.

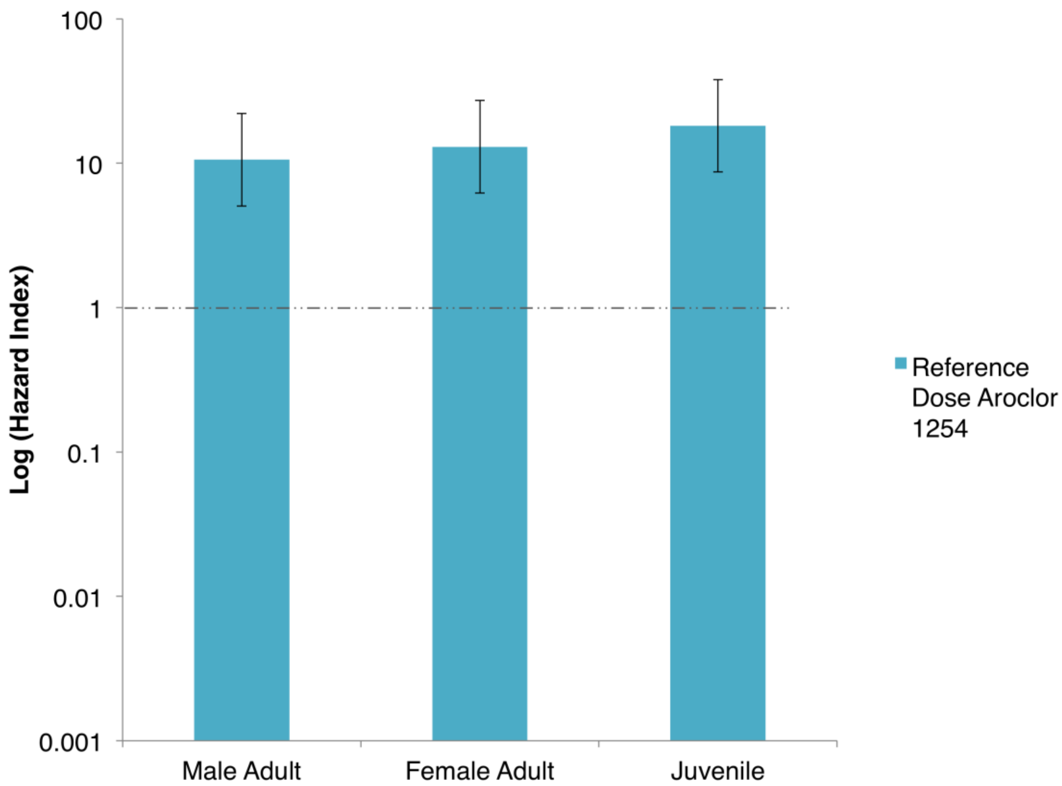

Figure 4.84 **Log-transformed hazard indices for polychlorinated biphenyls (PCBs) in three different life stages of resident killer whales, based on the reference dose for Aroclor 1254. The dashed-dotted line shows when the hazard index is equal to one.**

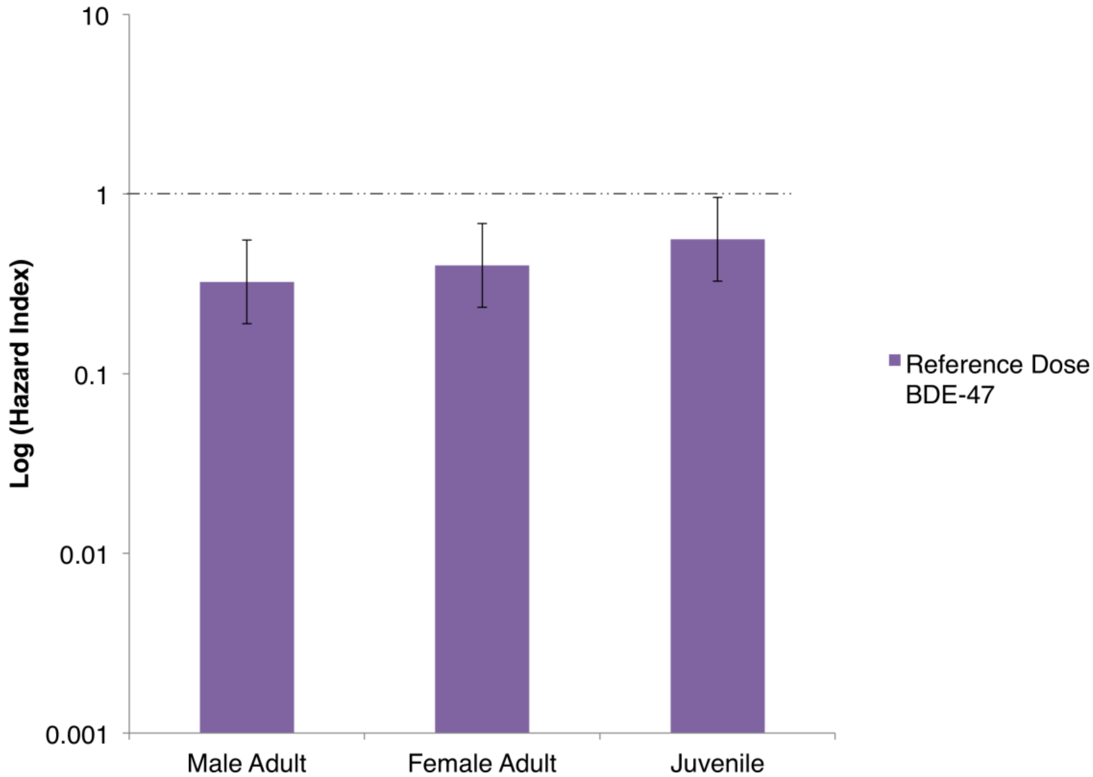

Figure 4.85 **Log-transformed hazard indices for polybrominated diphenyl ethers (PBDEs) in three different life stages of resident killer whales, based on the reference dose for BDE-47. The dashed-dotted line shows when the hazard index is equal to one.**

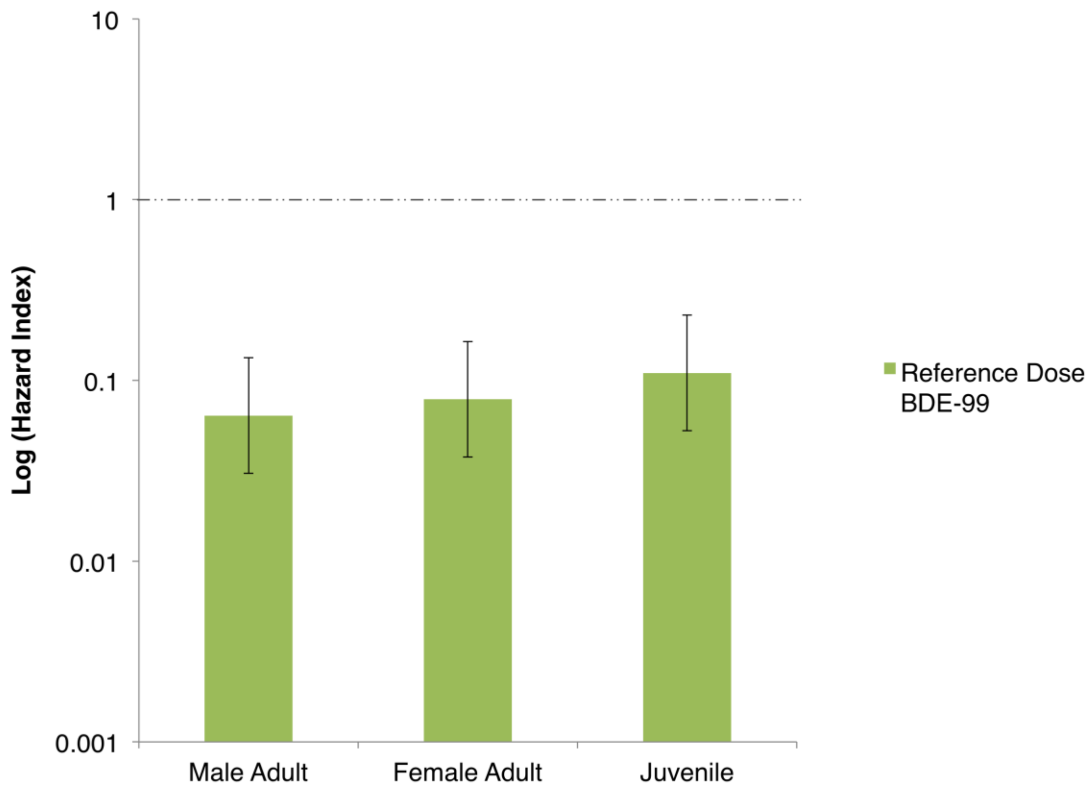

Figure 4.86 Log-transformed hazard indices for polybrominated diphenyl ethers (PBDEs) in three different life stages of resident killer whales, based on the reference dose for BDE-99. The dashed-dotted line shows when the hazard index is equal to one.

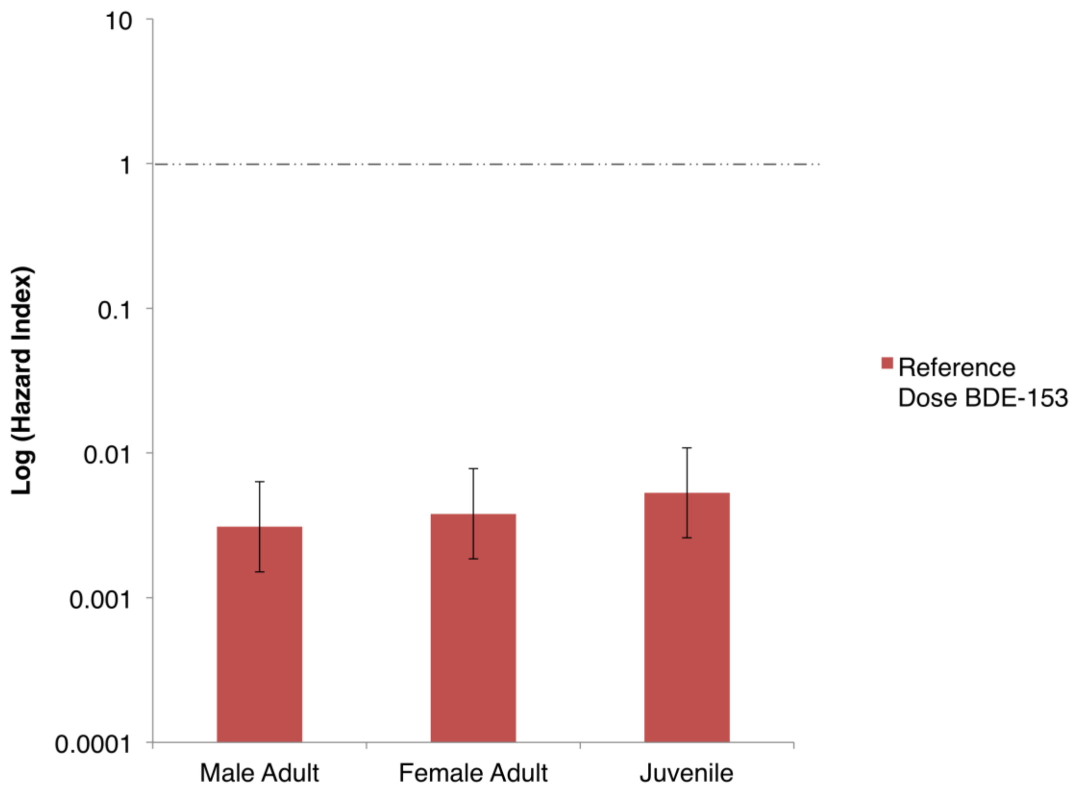

Figure 4.87 **Log-transformed hazard indices for polybrominated diphenyl ethers (PBDEs) in three different life stages of resident killer whales, based on the reference dose for BDE-153. The dashed-dotted line shows when the hazard index is equal to one.**

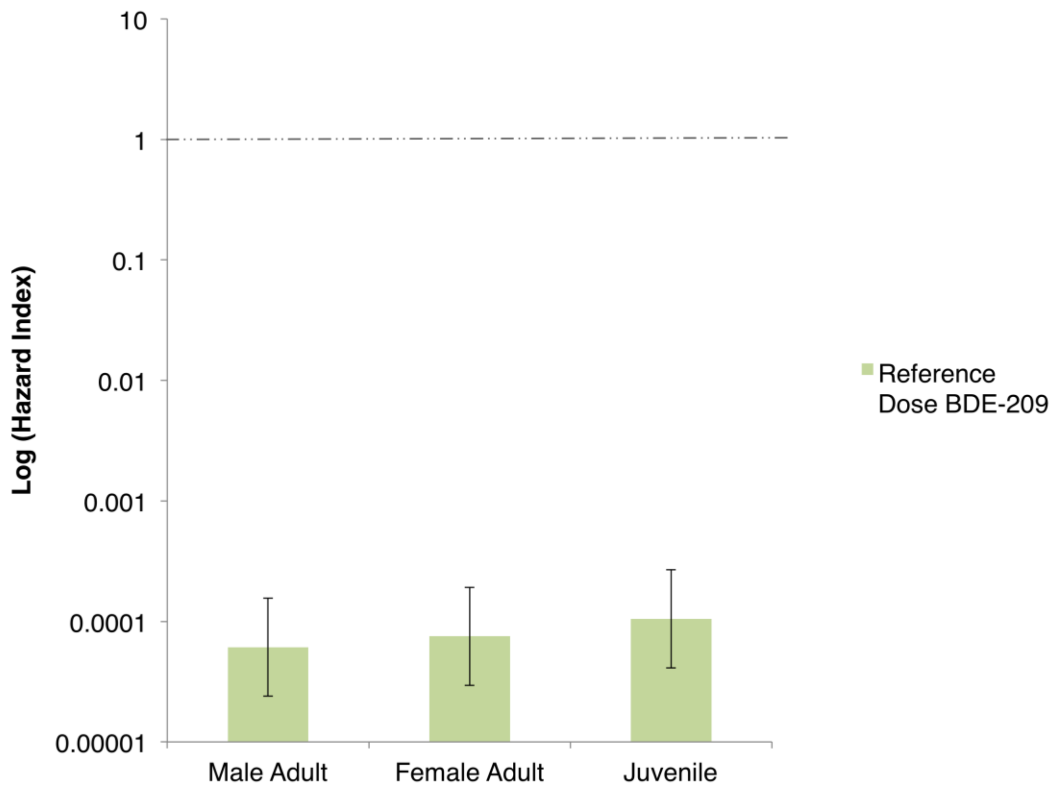

Figure 4.88 Log-transformed hazard indices for polybrominated diphenyl ethers (PBDEs) in three different life stages of resident killer whales, based on the reference dose for BDE-209. The dashed-dotted line shows when the hazard index is equal to one.

Table 4.32 Hazard indices for polychlorinated biphenyls (PCBs) and four polybrominated diphenyl ether congeners (BDE-47, BDE-99, BDE-153 and BDE-209) in different life stages of resident killer whales. Estimated daily exposure rates that were used for the hazard index calculations are given. The reference dose for negative health effects for each contaminant is also shown ($mg \cdot kg^{-1} \cdot day^{-1}$).

Contaminant	Sex & Life Stage	Estimated Daily Exposure Rate ($mg \cdot kg^{-1} \cdot day^{-1}$)	Contaminant for Reference Dose	Reference Dose ($mg \cdot kg^{-1} \cdot day^{-1}$)	Hazard Index	Upper Standard Deviation	Lower Standard Deviation
PCBs	Male Adult	2.11E-04	Aroclor 1016	7.00E-05	3.02E+00	6.31E+00	1.44E+00
PCBs	Female Adult	2.60E-04	Aroclor 1016	7.00E-05	3.72E+00	7.77E+00	1.78E+00
PCBs	Juvenile	3.63E-04	Aroclor 1016	7.00E-05	5.19E+00	1.09E+01	2.48E+00
PCBs	Male Adult	2.11E-04	Aroclor 1254	2.00E-05	1.06E+01	2.21E+01	5.05E+00
PCBs	Female Adult	2.60E-04	Aroclor 1254	2.00E-05	1.30E+01	2.72E+01	6.22E+00
PCBs	Juvenile	3.63E-04	Aroclor 1254	2.00E-05	1.82E+01	3.80E+01	8.69E+00
BDE-47	Male Adult	3.25E-05	BDE-47	1.00E-04	3.25E-01	5.56E-01	1.90E-01
BDE-47	Female Adult	4.00E-05	BDE-47	1.00E-04	4.00E-01	6.84E-01	2.34E-01
BDE-47	Juvenile	5.59E-05	BDE-47	1.00E-04	5.59E-01	9.56E-01	3.27E-01
BDE-99	Male Adult	3.76E-05	BDE-99	1.00E-04	6.39E-02	1.33E-01	3.06E-02
BDE-99	Female Adult	4.63E-05	BDE-99	1.00E-04	7.86E-02	1.64E-01	3.77E-02
BDE-99	Juvenile	6.47E-05	BDE-99	1.00E-04	1.10E-01	2.29E-01	5.27E-02
BDE-153	Male Adult	8.83E-06	BDE-153	2.00E-04	3.09E-03	6.32E-03	1.51E-03
BDE-153	Female Adult	1.09E-05	BDE-153	2.00E-04	3.80E-03	7.78E-03	1.86E-03
BDE-153	Juvenile	1.52E-05	BDE-153	2.00E-04	5.31E-03	1.09E-02	2.59E-03

Contaminant	Sex & Life Stage	Estimated Daily Exposure Rate (mg·kg^{-1}·day^{-1})	Contaminant for Reference Dose	Reference Dose (mg·kg^{-1}·day^{-1})	Hazard Index	Upper Standard Deviation	Lower Standard Deviation
BDE-209	Male Adult	8.42E-07	BDE-209	7.00E-03	6.11E-05	1.56E-04	2.39E-05
BDE-209	Female Adult	1.04E-06	BDE-209	7.00E-03	7.51E-05	1.92E-04	2.94E-05
BDE-209	Juvenile	1.45E-06	BDE-209	7.00E-03	1.05E-04	2.68E-04	4.11E-05

Carcinogenic risk estimates in the different life stages of resident killer whales were calculated for PCBs (Figure 4.89; Table 4.33). Four out of 1000 male adult killer whales, five out of 1000 female adult killer whales, and seven out of 1000 juvenile killer whales were potentially experiencing cancerous effects due to their daily exposure rate to PCBs.

Overall, both the hazard indices and the carcinogenic risk estimates showed that PCBs are possibly impacting the health of resident killer whales.

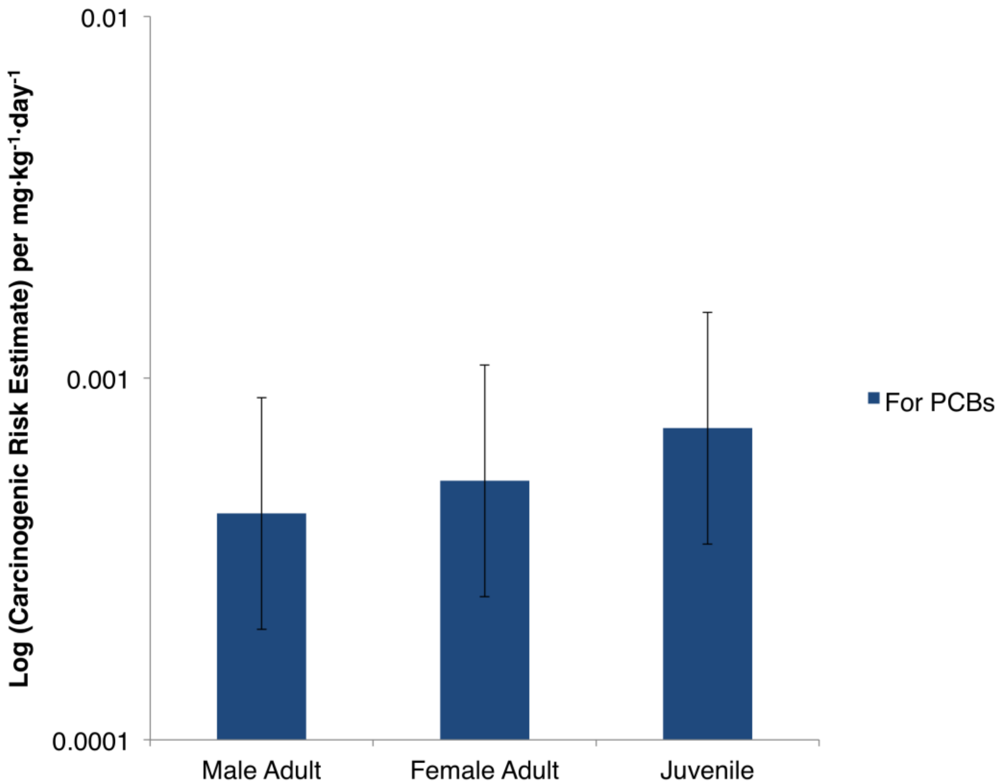

Figure 4.89 Log-transformed carcinogenic risk estimates for polychlorinated biphenyls (PCBs) in three different life stages of resident killer whales, based on a carcinogenic risk value from oral exposure to PCBs of 2.00 mg·kg^{-1}·day^{-1}.

Table 4.33 Carcinogenic risk estimates for polychlorinated biphenyls (PCBs) in male adult, female adult and juvenile killer whales. The estimated daily exposure rates ($mg \cdot kg^{-1} \cdot day^{-1}$) used for the carcinogenic risk estimate calculation are given along with the carcinogenic risk values (per $mg \cdot kg^{-1} \cdot day^{-1}$).

Contaminant	Sex & Life Stage	Estimated Daily Exposure Rate ($mg \cdot kg^{-1} \cdot day^{-1}$)	Contaminant for Carcinogenic Risk from Oral Exposure	Carcinogenic Risk from Oral Exposure (per $mg \cdot kg^{-1} \cdot day^{-1}$)	Carcinogenic Risk Estimate	Upper Standard Deviation	Lower Standard Deviation
PCBs	Male Adult	2.11E-04	PCBs	2.00E+00	4.23E-04	8.84E-04	2.02E-04
PCBs	Female Adult	2.60E-04	PCBs	2.00E+00	5.20E-04	1.09E-03	2.49E-04
PCBs	Juvenile	3.63E-04	PCBs	2.00E+00	7.27E-04	1.52E-03	3.48E-04

4.3.4. Risk-based List

A risk-based list of the contaminants explored in this study was compiled to advise the DFO and other interested parties concerning which contaminants were the greatest health risks to Salish Sea killer whales (Table 4.34). All the risk assessment outcomes for this study unanimously showed the legacy contaminant, PCBs, to be the main contaminant of concern to the health of Salish Sea killer whales. This list was based on contaminant comparisons and risk evaluations, including cumulative risk distributions, BMF values, daily exposure rates, hazard indices, and carcinogenic risk estimates.

Table 4.34 A risk-based list of nine contaminants, indicating from greatest to least concern to the health of Salish Sea killer whales. The contaminants explored were total polychlorinated biphenyls (ΣPCBs), total polybrominated diphenyl ethers (ΣPBDEs), total isomers of hexabromocyclododecane (ΣHBCDDs), Dechlorane, 1,2-dibromobenzene (1,2-DiBB), pentabromotoluene (PBT), tetrabromo-o-chlorotoluene (TBCT), Dechlorane Plus and Dechlorane 602 (Dec 602).

Contaminant	Ranking of Health Risk to Killer Whale Health
ΣPCBs	Contaminants of Highest Concern
ΣPBDEs	Contaminants of Second-highest Concern
HBCDD	
Dechlorane (Mirex)	
Dechlorane Plus	
Dec 602	Contaminants of Least Concern
1,2-DiBB	
PBT	
TBCT	

PBDEs may not be the contaminant of greatest concern, but they must be monitored in killer whales and their food sources over time. Such monitoring must be carried out to prevent PBDEs from surpassing the risk to the health of killer whales that is currently posed by PCBs. HBCDDs and the other flame-retardants that were investigated were not of great concern to the health of Salish Sea killer whales. Even though HBCDDs and the other emerging flame-retardants were of least concern to the health of the Salish Sea killer whales, the risk-based list ranks the chemicals from the greatest to the least concern. This is based on the geometric mean concentrations of the contaminants found in the killer whales on a molar bases (see Table 4.3). The geometric mean concentrations were used to rank the risk of these contaminants, as there were no known toxicity reference values of these contaminants in marine mammals at the time of this present study. HBCDD was found to be the contaminant of most concern to the health of the killer whales among the emerging flame-retardants. TBCT was found to be the least concern to the health of the killer whales among the emerging flame-retardants. This study serves as a baseline for these emerging contaminants and as a reference for future studies, both of which are important due to the current lack of data concerning these pollutants in relation to the health of killer whales.

Chapter 5. Conclusion

This study attempted to measure the concentrations of PCBs, PBDEs, and several other emerging contaminants present in Salish Sea killer whales. A risk assessment approach was used to determine which contaminant was of greatest concern to the health of the killer whales. New samples were collected from killer whales and Chinook salmon to determine their concentrations of PCBs, PBDEs, and emerging pollutants (i.e., ΣHBCDDs, Dechlorane, DP, Dec 602, PBT, 1,2-DiBB, and TBCT). This is the first time that concentrations of ΣHBCDDs, Dechlorane, DP, Dec 602, PBT, 1,2-DiBB, and TBCT have been measured in killer whales, offering a foundation for future implications of these contaminants.

This study determined that concentrations of ΣPCBs have not changed significantly over time in either killer whales or Chinook salmon, their main prey. Concentrations of ΣPBDEs, in contrast, appear to have been increasing significantly over time in killer whales. There were insufficient data concerning concentrations of ΣPBDEs in Chinook salmon to allow a conclusion of changes in concentration over time, highlighting a gap in data concerning contaminant measurements in Chinook salmon and indicating a need for the collection of more contaminant data from Chinook salmon in the future. This is important because Chinook salmon comprise a significant portion of the diet of resident killer whales, and understanding the diet of resident killer whales will help in gaining a better understanding of their health.

The age of killer whales, largely regardless of ecotype and sex, was found to not be a sampling artefact. The only ecotype for which age possibly affected the data set was female Bigg’s killer whales, when testing for changes in concentrations of PBDEs over time. The significant changes in concentrations of PBDEs over time in female Bigg’s killer whales found in this study were therefore considered to be inaccurate.

When comparing all explored contaminants to PCBs, it was determined that PCBs remain the main contaminant of concern for the health of Salish Sea killer whales. Interestingly, PBDEs were found to be increasingly more toxic in killer whales over time. This was true in Chinook salmon as well, where fractions of contaminant concentrations above the Canadian federal quality guidelines for wildlife consumption of PBDEs were considered to be more toxic than those of PCBs.

Of the killer whales sampled for PCBs, 96.6% were found to contain levels above the average marine mammal toxicity threshold value for PCBs. In contrast, 34.4% of killer whales sampled for PBDEs contained levels above the marine mammal toxicity threshold for PBDEs. In Chinook salmon, 31.6% of samples were found to contain concentrations of PCBs that were above the present study's reworked tissue residue guideline of 0.688 $mg \cdot kg^{-1}$ ww. For PBDEs, however, 50.0% of Chinook salmon samples contained levels above the reworked Canadian federal quality guideline for PBDEs.

Newly calculated BMF values for PCBs, PBDEs, and HBCDDs will help with predicting contaminant concentrations in killer whales by investigating contaminants in the diet of killer whales. Estimated daily exposure rates to PCBs indicated that juvenile killer whales experienced the highest risk of health impacts due to ingestion of PCBs through a diet of Chinook salmon.

Hazard indices related that PCBs were hazardous to the health of killer whales in all life stages. Four out 1000 adult male killer whales, five out of 1000 adult female killer whales, and seven out 1000 juvenile killer whales were found to be potentially experiencing cancerous effects due to PCBs.

A risk-based list was compiled by considering all the outcomes assessed in the present study, and overall PCBs were found to be the main contaminant of concern to the health of Salish Sea killer whales. PBDEs were found to be a rising concern to the health of the Salish sea killer whales, however, and should therefore be monitored. Other emerging contaminants, including HBCDDs, were not main concerns to the health of killer whales, in comparison to PCBs.

In the future, PCBs and PBDEs should continuously be monitored in killer whales, as concentrations could change over time. Sources inputting PCBs and PBDEs into the marine ecosystem must also be determined and mitigated. PCBs and PBDEs should also be monitored in Chinook salmon, necessitating the collection of more Chinook salmon data. This study showed that resident killer whales' diet of Chinook salmon is a key component to understanding the concentrations of contaminants in killer whales. Collecting and sampling Chinook salmon not only alleviates the difficulties involved in collecting blubber samples from killer whales, but also grants a more holistic understanding of how contaminants may be affecting the marine ecosystem.

This study will help in the development of frameworks for the second stage of the recovery strategy for resident killer whales in the Action Plan implemented by the Department of Fisheries and Oceans (DFO). This study will also help inform remediation of contaminated sites, and other risked-based assessments for dredge and disposal options in British Columbia’s coastal waters.

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Appendix A.

List of Chemicals Analyzed

Table A.1 List of chemicals measured within Salish Sea killer whales sampled in 2015 and Chinook salmon in 2014. The full name and abbreviation for each chemical, as well as if the chemical was detected in all samples, are given.

Contaminants Measured	Abbreviation	Detected
Polychlorinated biphenyls	PCBs	Yes
Polybrominated diphenyl ethers	PBDEs	Yes
Hexabromocyclododecane	HBCDs or HBCDDs	Yes
Dechlorane	Mirex	Yes
Dechlorane Plus	DDC-CO	Yes
Dechlorane 602	Dec 602	Yes
Dechlorane 603	Dec 603	No
Dechlorane 604	Dec 604	No
Hexachlorocyclopentadienyldibromocyclooctane	HCDBCO or DBHCTD	No
Allyl 2,4,6-tribromophenyl ether	ATE	No
2-bromoallyl 2,4,6-tribromophenyl ether	BATE	No
2,3-dibromopropyl-2,4,6-tribromophenyl ether	DPTE	No
1,2-bis(2,4,6-tribromophenoxy)ethane	BTBPE	No
2-ethyl-1-hexyl 2,3,4,5-tetrabromobenzoate	EHTBB	No
1,2-dibromo-4-(1,2-dibromoethyl)-cyclohexane	TBECH	No
Hexabromobenzene	HBB	No
1,2,3,4,5-pentabromo-benzene	PBBZ	No
1,2,4,5/1,2,3,5-tetrabromobenzene	1,2,4,5/1,2,3,5-TBB	No
1,2,4-tribromobenzene	1,2,4-TriBB	No
1,2-dibromobenzene	1,2-DiBB or 1,2-DBB	Yes
1,4-dibromobenzene	1,4-DiBB or 1,4-DBB	No
Pentabromotoluene	PBT	Yes
Pentabromoethylbenzene	PBEB	No

Pentabromobenzylbromide	PBBB	No
2,3,5,6-tetrabromo-*p*-xylene	pTBX	No
Tetrabromo-o-chlorotoluene	TBCT	Yes

Appendix B.
Supplementary Data Files

Description:

Two accompanying csv files contain the analyzed concentrations for all detected and non-detected contaminants measured in all killer whales sampled in 2015 and all Chinook salmon sampled in 2014. These csv files contain the original lab results. The concentrations are expressed in pg/g wet weight and are not blank corrected. The five tabs that are found in both files are listed below with their descriptions:

- "PCBs" – This tab contains the individual concentrations of each PCB congener for all samples.
- "PBDEs" – This tab contains the individual concentrations for each PBDE congener for all samples.
- "HBCDDs" – This tab contains the individual concentrations for each HBCDD isomer for all samples.
- "CFRs" – This tab contains the individual concentrations for each chlorinated or brominated flame retardant for all samples.
- "OCPs" – This tab contains the individual concentrations for each organochlorine pesticide for all samples.

Files names:

I. GuyJayda_MRM699_datafile_KW2016.csv

II. GuyJayda_MRM699_datafile_CS2016.csv

Appendix C.

Analysis Methods for All Samples

Table C.1 All analyses included in this study. Both analyses from the present study and compiled past analyses are included.

Animal (Species)	Year	Contaminant Analyzed	Sub-analysis	Detection?	Methods	Number of Congeners Analyzed
Killer Whales *(Orcinus orca)*	1993 [62]	PCBs	No	Yes	HRGC/HRMS	205
	1993 [8]	PBDEs	Yes	Yes	HRGC/HRMS	37
	1994 [8]	PBDEs	Yes	Yes	HRGC/HRMS	37
	1995 [8]	PBDEs	Yes	Yes	HRGC/HRMS	37
	1996 [62]	PCBs	No	Yes	HRGC/HRMS	205
	1996 [8]	PBDEs	Yes	Yes	HRGC/HRMS	37
	1997 [8]	PBDEs	Yes	Yes	HRGC/HRMS	37
	2000 [62]	PCBs	No	Yes	HRGC/HRMS	205
	2002 [62]	PCBs	No	Yes	HRGC/HRMS	205
	2003 [62]	PCBs	Yes	Yes	HRGC/HRMS	205
	2003 [62]	PBDEs	Yes	Yes	HRGC/HRMS	78
	2004 [62]	PCBs	Yes	Yes	HRGC/HRMS	205
	2004 [32]	PCBs	Yes	Yes	LC GC/MS	45
	2004 [62]	PBDEs	Yes	Yes	HRGC/HRMS	78
	2006 [32]	PCBs	Yes	Yes	LC GC/MS	45
	2007 [62]	PCBs	Yes	Yes	HRGC/HRMS	205
	2007 [33]	PCBs	Yes	Yes	LC GC/MS	45
	2007 [62]	PBDEs	Yes	Yes	HRGC/HRMS	78
	2007 [33]	PBDEs	Yes	Yes	LC GC/MS	10
	2008 [62]	PCBs	Yes	Yes	HRGC/HRMS	205
	2008 [62]	PBDEs	Yes	Yes	HRGC/HRMS	78
	2009 [62]	PCBs	Yes	Yes	HRGC/HRMS	205
	2009 [62]	PBDEs	Yes	Yes	HRGC/HRMS	78

Animal (Species)	Year	Contaminant Analyzed	Sub-analysis	Detection?	Methods	Number of Congeners Analyzed
	2015*	PCBs	Yes	Yes	HRGC/HRMS	209
	2015*	PBDEs	Yes	Yes	HRGC/HRMS	46
	2015*	HBCDDs	Yes	Yes	LC MS/MS	3 isomers
	2015*	Dechlorane	Yes	Yes	LR GC/MS	n/a
	2015*	Dechlorane Plus	Yes	Yes	LR GC/MS	Anti + Syn
	2015*	Dec 602	Yes	Yes	LR GC/MS	n/a
	2015*	Dec 603	Yes	No	LR GC/MS	n/a
	2015*	Dec 604	Yes	No	LR GC/MS	n/a
	2015*	HCDBCO	Yes	No	LR GC/MS	n/a
	2015*	ATE	Yes	No	LR GC/MS	n/a
	2015*	BATE	Yes	No	LR GC/MS	n/a
	2015*	DPTE	Yes	No	LR GC/MS	n/a
	2015*	BTBPE	Yes	No	LR GC/MS	n/a
	2015*	EHTBB	Yes	No	LR GC/MS	n/a
	2015*	Total TBECH	Yes	No	LR GC/MS	n/a
	2015*	HBB	Yes	No	LR GC/MS	n/a
	2015*	PBBZ	Yes	No	LR GC/MS	n/a
	2015*	1,2,4,5/1,2,3,5 -TBB	Yes	No	LR GC/MS	n/a
	2015*	1,2,4-TriBB	Yes	No	LR GC/MS	n/a
	2015*	1,2-DiBB	Yes	Yes	LR GC/MS	n/a
	2015*	1,4-DiBB	Yes	No	LR GC/MS	n/a
	2015*	PBT	Yes	Yes	LR GC/MS	n/a
	2015*	PBEB	Yes	No	LR GC/MS	n/a
	2015*	PBBB	Yes	No	LR GC/MS	n/a
	2015*	pTBX	Yes	No	LR GC/MS	n/a
	2015*	TBCT	Yes	Yes	LR GC/MS	n/a
Chinook	2000[3]	PCBs	Yes	Yes	HRGC/HRMS	209

Animal (Species)	Year	Contaminant Analyzed	Sub-analysis	Detection?	Methods	Number of Congeners Analyzed
Salmon (*Oncorhynchus tshawytscha*)	2002**	PBDEs	Yes	Yes	HRGC/HRMS	44
	2014*	PCBs	Yes	Yes	HRGC/HRMS	209
	2014*	PBDEs	Yes	Yes	HRGC/HRMS	46
	2014*	HBCDDs	Yes	Yes	LC MS/MS	3 isomers
	2014*	Dechlorane	Yes	No	LR GC/MS	n/a
	2014*	Dechlorane Plus	Yes	No	LR GC/MS	Anti + Syn
	2014*	Dec 602	Yes	No	LR GC/MS	n/a
	2014*	Dec 603	Yes	No	LR GC/MS	n/a
	2014*	Dec 604	Yes	No	LR GC/MS	n/a
	2014*	HCDBCO	Yes	No	LR GC/MS	n/a
	2014*	ATE	Yes	No	LR GC/MS	n/a
	2014*	BATE	Yes	No	LR GC/MS	n/a
	2014*	DPTE	Yes	No	LR GC/MS	n/a
	2014*	BTBPE	Yes	No	LR GC/MS	n/a
	2014*	EHTBB	Yes	No	LR GC/MS	n/a
	2014*	Total TBECH	Yes	No	LR GC/MS	n/a
	2014*	HBB	Yes	No	LR GC/MS	n/a
	2014*	PBBZ	Yes	No	LR GC/MS	n/a
	2014*	1,2,4,5/1,2,3,5 -TBB	Yes	No	LR GC/MS	n/a
	2014*	1,2,4-TriBB	Yes	No	LR GC/MS	n/a
	2014*	1,2-DiBB	Yes	No	LR GC/MS	n/a
	2014*	1,4-DiBB	Yes	No	LR GC/MS	n/a
	2014*	PBT	Yes	No	LR GC/MS	n/a
	2014*	PBEB	Yes	No	LR GC/MS	n/a
	2014*	PBBB	Yes	No	LR GC/MS	n/a
	2014*	pTBX	Yes	No	LR GC/MS	n/a
	2014*	TBCT	Yes	No	LR GC/MS	n/a

*present study, **unpublished analysis from 2002 done at Axys Analytical Ltd in Sidney, British Columbia, Canada

Appendix D.

Student T-tests of Detection Limit Methods

For all samples that possessed concentrations below the detection limit (DL), the detection limit value was taken for the concentration calculations. Student t-tests were conducted to compare different detection limit methods. The other detection limit methods tested included recording the DL as zero or as 50% of the DL if the concentration was below the DL. The majority of the student t-tests that were conducted found no significant difference between the detection limit methods. The only situation where there was a significant difference in the detection limit methods was for the concentrations of HCBDDs in Chinook salmon, which occurred because two of the isomers of HBCDD were undetected. The detection limit value was still taken for this situation.

Table D.1 Results of student t-tests investigating if there were significant differences between methods of determining detection limits (DL). These tests were conducted for all contaminants analyzed in killer whales sampled for this present study. Differences between means, standard deviations and p-values are given.

Detection Limit (DL) - Level 1	Detection Limit (DL) - Level 2	Animal	Contaminant	N	Difference	Standard Deviation	p-value
DL=DL	DL=0	Killer Whales	PCB	9	2.45E+03	5.02E+06	1.00E+00
DL=0.5DL	DL=0	Killer Whales	PCB	9	1.22E+03	5.02E+06	1.00E+00
DL=DL	DL=0.5DL	Killer Whales	PCB	9	1.22E+03	5.02E+06	1.00E+00
DL=DL	DL=0	Killer Whales	PBDE	9	3.29E+03	6.44E+05	9.96E-01
DL=0.5DL	DL=0	Killer Whales	PBDE	9	1.65E+03	6.44E+05	9.98E-01
DL=DL	DL=0.5DL	Killer Whales	PBDE	9	1.65E+03	6.44E+05	9.98E-01
DL=DL	DL=0	Killer Whales	HBCDD	9	1.51E+01	8.25E+00	7.96E-02

DL=DL	DL=0.5DL	Killer Whales	HBCDD	9	7.55E+00	8.25E+00	3.69E-01
DL=0.5DL	DL=0	Killer Whales	HBCDD	9	7.55E+00	8.25E+00	3.69E-01
DL=DL	DL=0	Killer Whales	1,2-DiBB	9	0.2666667	8.98E+00	9.77E-01
DL=DL	DL=0.5DL	Killer Whales	1,2-DiBB	9	0.1333333	8.98E+00	9.88E-01
DL=0.5DL	DL=0	Killer Whales	1,2-DiBB	9	0.1333333	8.98E+00	9.88E-01
DL=0.5DL	DL=0	Killer Whales	Dec 602	9	0	4.42E-01	1.00E+00
DL=DL	DL=0	Killer Whales	Dec 602	9	0	4.42E-01	1.00E+00
DL=DL	DL=0.5DL	Killer Whales	Dec 602	9	0	4.42E-01	1.00E+00
DL=0.5DL	DL=0	Killer Whales	Dechlorane	9	0	1.46E+01	1.00E+00
DL=DL	DL=0	Killer Whales	Dechlorane	9	0	1.46E+01	1.00E+00
DL=DL	DL=0.5DL	Killer Whales	Dechlorane	9	0	1.46E+01	1.00E+00
DL=DL	DL=0	Killer Whales	Dechlorane Plus	9	1.002333	9.32E-01	2.93E-01
DL=0.5DL	DL=0	Killer Whales	Dechlorane Plus	9	0.501167	9.32E-01	5.96E-01
DL=DL	DL=0.5DL	Killer Whales	Dechlorane Plus	9	0.501167	9.32E-01	5.96E-01
DL=DL	DL=0	Killer Whales	PBT	9	0.0444444	1.48E+00	9.76E-01
DL=DL	DL=0.5DL	Killer Whales	PBT	9	0.0222222	1.48E+00	9.88E-01
DL=0.5DL	DL=0	Killer Whales	PBT	9	0.0222222	1.48E+00	9.88E-01
DL=0.5DL	DL=0	Killer Whales	TBCT	9	0	1.72E+00	1.00E+00
DL=DL	DL=0	Killer Whales	TBCT	9	0	1.72E+00	1.00E+00
DL=DL	DL=0.5DL	Killer Whales	TBCT	9	0	1.72E+00	1.00E+00

Table D.2 **Results of student t-tests investigating if there were significant differences between methods of determining detection limits (DL). These tests were conducted for all contaminants analyzed in the Chinook salmon sampled for this present study. Differences between means, standard deviations and p-values are given.**

Detection Limit (DL) - Level 1	Detection Limit (DL) - Level 2	Animal	Contaminant	N	Difference	Standard Deviation	p-value
DL=DL	DL=0	Chinook Salmon	PCB	7	1.09E+01	3.43E+03	9.98E-01
DL=0.5DL	DL=0	Chinook Salmon	PCB	7	5.57E+00	3.43E+03	9.99E-01
DL=DL	DL=0.5DL	Chinook Salmon	PCB	7	5.29E+00	3.43E+03	9.99E-01
DL=DL	DL=0	Chinook Salmon	PBDE	7	4.97E+01	6.34E+02	9.38E-01
DL=0.5DL	DL=0	Chinook Salmon	PBDE	7	2.49E+01	6.34E+02	9.69E-01
DL=DL	DL=0.5DL	Chinook Salmon	PBDE	7	2.49E+01	6.34E+02	9.69E-01
DL=DL	DL=0	Chinook Salmon	HBCDD	7	1.99E-01	3.19E-02	<0.0001
DL=DL	DL=0.5DL	Chinook Salmon	HBCDD	7	9.95E-02	3.19E-02	5.90E-03
DL=0.5DL	DL=0	Chinook Salmon	HBCDD	7	9.95E-02	3.19E-02	5.90E-03

Appendix E.
List of Contaminant Concentrations

Table E.1 Concentrations of total polychlorinated biphenyls (ΣPCB) measured in the blubber of killer whales from 1993 to 2015.

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg \cdot kg^{-1}{}_{lw}$)
Ross et al. 2013	A27	N. Res	M	22	1993	205	64.3	2.42E+00
Ross et al. 2013	A43	N. Res	F	12	1993	205	64.3	7.41E+00
Ross et al. 2013	A23	N. Res	F	46	1993	205	64.3	2.58E+00
Ross et al. 2013	A60	N. Res	M	1	1993	205	64.3	1.31E+01
Ross et al. 2013	A24	N. Res	F	26	1993	205	64.3	4.79E-01
Ross et al. 2013	A6	N. Res	M	29	1993	205	64.3	1.79E+01
Ross et al. 2013	B1	N. Res	M	42	1993	205	64.3	6.90E+00
Ross et al. 2013	B2	N. Res	M	41	1993	205	64.3	2.69E+01
Ross et al. 2013	A54	N. Res	F	4	1993	205	64.3	9.96E+00
Ross et al. 2013	A11	N. Res	F	35	1993	205	64.3	1.04E+00
Ross et al. 2013	A13	N. Res	M	15	1993	205	64.3	2.12E+01
Ross et al. 2013	A35	N. Res	F	19	1993	205	64.3	1.68E+00
Ross et al. 2013	A48	N. Res	F	10	1993	205	64.3	1.07E+01
Ross et al. 2013	A52	N. Res	F	6	1993	205	64.3	3.39E+01
Ross et al. 2013	A56	N. Res	F	3	1993	205	64.3	9.80E+00
Ross et al. 2013	A59	N. Res	M	1	1993	205	64.3	1.08E+01
Ross et al. 2013	A9	N. Res	F	53	1993	205	64.3	2.40E+01
Ross et al. 2013	I2	N. Res	F	57	1996	205	64.3	9.45E+00
Ross et al. 2013	C5	N. Res	F	71	1996	205	64.3	2.55E+01
Ross et al. 2013	B12	N. Res	M	12	1996	205	64.3	2.00E+01
Ross et al. 2013	B13	N. Res	M	9	1996	205	64.3	2.79E+01
Ross et al. 2013	A5	N. Res	M	39	1996	205	64.3	3.82E+01
Ross et al. 2013	I5	N. Res	M	42	1996	205	64.3	3.77E+01

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg{\cdot}kg^{-1}{}_{lw}$)
Ross et al. 2013	R6	N. Res	M	42	1996	205	64.3	4.96E+01
Ross et al. 2013	H4	N. Res	M	22	1996	205	64.3	2.20E+01
Ross et al. 2013	C10	N. Res	F	25	1996	205	64.3	6.90E+00
Ross et al. 2013	A42	N. Res	F	16	1996	205	64.3	1.54E+01
Ross et al. 2013	A57	N. Res	F	5	1996	205	64.3	1.09E+02
Ross et al. 2013	C17	N. Res	M	11	2000	205	64.3	6.89E+00
Ross et al. 2013	A70	N. Res	F	1	2000	205	64.3	4.12E+00
Ross et al. 2013	A69	N. Res	F	4	2000	205	64.3	1.06E+01
Ross et al. 2013	I85	N. Res	M	2	2000	205	64.3	5.75E+00
Ross et al. 2013	I51	N. Res	F	14	2000	205	64.3	7.85E+00
Ross et al. 2013	G51	N. Res	F	8	2000	205	64.3	1.57E+01
Ross et al. 2013	I80	N. Res	F	3	2000	205	64.3	1.94E+01
Ross et al. 2013	I15	N. Res	F	48	2000	205	64.3	1.86E+00
Ross et al. 2013	I63	N. Res	F	10	2000	205	64.3	1.79E+01
Ross et al. 2013	C20	N. Res	M	7	2000	205	64.3	5.46E+00
Ross et al. 2013	D12	N. Res	F	20	2002	205	64.3	1.51E+00
Ross et al. 2013	C18	N. Res	M	11	2002	205	64.3	6.28E+00
Ross et al. 2013	I 21	N. Res	F	23	2002	205	64.3	2.45E+00
Ross et al. 2013	D13	N. Res	F	18	2002	205	64.3	3.48E+00
Ross et al. 2013	I 52	N. Res	M	16	2002	205	64.3	8.24E+00
Ross et al. 2013	I 50	N. Res	F	20	2002	205	64.3	3.76E+00
Ross et al. 2013	D 09	N. Res	F	31	2003	205	64.3	6.37E+00
Ross et al. 2013	A55	N. Res	M	13	2003	205	64.3	2.60E+00
Ross et al. 2013	A60	N. Res	M	11	2003	205	64.3	1.53E+01
Ross et al. 2013	I68	N. Res	F	11	2003	205	64.3	7.57E+00
Ross et al. 2013	A33	N. Res	M	32	2003	205	64.3	1.12E+01
Ross et al. 2013	I67	N. Res	M	12	2003	205	64.3	2.37E+01
Ross et al. 2013	I42	N. Res	M	20	2003	205	64.3	7.15E+00
Ross et al. 2013	A74	N. Res	M	3	2003	205	64.3	6.83E+00

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg \cdot kg^{-1}{}_{lw}$)
Ross et al. 2013	A62	N. Res	F	10	2003	205	64.3	3.54E+00
Ross et al. 2013	R28	N. Res	M	15	2003	205	64.3	3.48E+00
Ross et al. 2013	C16	N. Res	F	15	2004	205	64.3	6.65E+00
Ross et al. 2013	A71	N. Res	M	5	2004	205	64.3	3.27E+00
Ross et al. 2013	B14	N. Res	F	16	2007	205	64.3	1.37E+00
Ross et al. 2013	A71	N. Res	M	8	2007	205	64.3	8.18E+00
Ross et al. 2013	A61	N. Res	M	13	2007	205	64.3	9.78E+00
Ross et al. 2013	C22	N. Res	M	10	2007	205	64.3	8.52E+00
Ross et al. 2013	I64	N. Res	M	17	2007	205	64.3	4.47E+00
Ross et al. 2013	I80	N. Res	F	10	2007	205	64.3	2.50E+01
Ross et al. 2013	I46	N. Res	M	22	2007	205	64.3	7.22E-01
Ross et al. 2013	R31	N. Res	M	10	2007	205	64.3	2.12E+01
Ross et al. 2013	R30	N. Res	M	13	2007	205	64.3	6.59E+00
Ross et al. 2013	R43	N. Res	M	5	2007	205	64.3	1.16E+01
Ross et al. 2013	I35	N. Res	F	33	2007	205	64.3	1.21E+00
Ross et al. 2013	I110	N. Res	*	2	2007	205	64.3	5.14E+00
Ross et al. 2013	I46	N. Res	M	23	2008	205	64.3	1.53E+01
Ross et al. 2013	A67	N. Res	F	12	2008	205	64.3	5.33E+00
Ross et al. 2013	A78	N. Res	*	5	2008	205	64.3	2.78E+00
Ross et al. 2013	A51	N. Res	F	22	2008	205	64.3	1.11E+01
Ross et al. 2013	I62	N. Res	M	20	2008	205	64.3	8.36E+00
Ross et al. 2013	A84	N. Res	*	3	2008	205	64.3	9.30E+00
Ross et al. 2013	C24	N. Res	F	8	2008	205	64.3	6.14E+01
Ross et al. 2013	I110	N. Res	*	3	2008	205	64.3	1.07E+01
Ross et al. 2013	I64	N. Res	M	18	2008	205	64.3	6.81E+00
Ross et al. 2013	R28	N. Res	M	16	2008	205	64.3	9.11E+00
Ross et al. 2013	R43	N. Res	M	6	2008	205	64.3	1.33E+01
Ross et al. 2013	R30	N. Res	M	14	2008	205	64.3	1.02E+01
Ross et al. 2013	I42	N. Res	M	25	2008	205	64.3	1.46E+01

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg \cdot kg^{-1}{}_{lw}$)
Ross et al. 2013	I78	N. Res	M	11	2008	205	64.3	8.52E+00
Ross et al. 2013	I67	N. Res	M	17	2008	205	64.3	9.38E+00
Ross et al. 2013	A39	N. Res	M	33	2008	205	64.3	7.68E+00
Ross et al. 2013	A72	N. Res	F	7	2008	205	64.3	1.45E+01
Ross et al. 2013	A75	N. Res	F	7	2008	205	64.3	1.29E+01
Ross et al. 2013	A84	N. Res	*	3	2008	205	64.3	9.99E+00
Ross et al. 2013	A86	N. Res	*	2	2008	205	64.3	5.97E+00
Ross et al. 2013	A54	N. Res	F	19	2008	205	64.3	1.77E+00
Ross et al. 2013	A79	N. Res	*	4	2009	205	64.3	1.21E+01
Ross et al. 2013	R39	N. Res	F	8	2009	205	64.3	7.88E+00
Ross et al. 2013	R44	N. Res	*	5	2009	205	64.3	1.34E+01
Ross et al. 2013	R35	N. Res	F	11	2009	205	64.3	1.33E+01
Ross et al. 2013	R29	N. Res	F	15	2009	205	64.3	9.00E+00
Ross et al. 2013	off1	Offshore	*	*	1993	205	64.3	3.00E+01
Ross et al. 2013	off2	Offshore	*	*	1993	205	64.3	2.63E+01
Ross et al. 2013	O250	Offshore	*	*	2008	205	64.3	3.97E+01
Ross et al. 2013	J6	S. Res	M	37	1993	205	64.3	5.93E+00
Ross et al. 2013	J3	S. Res	M	40	1993	205	64.3	1.62E+02
Ross et al. 2013	J1	S. Res	M	46	1996	205	64.3	1.92E+02
Ross et al. 2013	J18	S. Res	M	20	1996	205	64.3	6.32E+01
Ross et al. 2013	J20	S. Res	F	16	1996	205	64.3	7.47E+01
Ross et al. 2013	J11	S. Res	F	41	1996	205	64.3	3.47E+01
Ross et al. 2013	J18	S. Res	M	23	2000	205	64.3	2.48E+02
Ross et al. 2013	L78	S. Res	M	15	2004	205	64.3	8.53E+00
Ross et al. 2013	L74	S. Res	M	18	2004	205	64.3	2.22E+01
Ross et al. 2013	L71	S. Res	M	18	2004	205	64.3	1.78E+01
Krahn et al. 2009	J22	S. Res	F	22	2007	45	28.4	4.60E+00
Krahn et al. 2009	J38	S. Res	M	4	2007	45	20.9	4.10E+01
Krahn et al. 2009	K7	S. Res	F	97	2007	45	28.5	1.20E+02

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg \cdot kg^{-1}{}_{lw}$)
Krahn et al. 2009	K13	S. Res	F	35	2007	45	22.0	8.90E+00
Krahn et al. 2009	K21	S. Res	M	35	2007	45	26.6	3.80E+01
Krahn et al. 2009	K34	S. Res	M	6	2007	45	22.3	3.90E+01
Krahn et al. 2009	K36	S. Res	F	4	2007	45	18.3	6.20E+01
Krahn et al. 2009	L21	S. Res	F	57	2007	45	18.7	5.50E+01
Krahn et al. 2009	L26	S. Res	F	51	2007	45	22.1	2.70E+01
Krahn et al. 2009	L67	S. Res	F	22	2007	45	29.2	4.30E+00
Krahn et al. 2009	L73	S. Res	M	21	2007	45	23.8	3.20E+01
Krahn et al. 2009	L87	S. Res	M	15	2007	45	25.6	2.40E+01
Krahn et al. 2007	J19	S. Res	F	27	2006	45	29.4	4.50E+01
Krahn et al. 2007	J39	S. Res	M	3	2006	45	40.9	3.40E+01
Krahn et al. 2007	J1	S. Res	M	55	2006	45	21.9	1.80E+02
Krahn et al. 2007	J27	S. Res	M	15	2006	45	30.4	7.40E+01
Krahn et al. 2007	L57	S. Res	M	29	2006	45	19.4	5.60E+01
Krahn et al. 2007	L71	S. Res	M	18	2004	45	9.60	3.60E+01
Krahn et al. 2007	L74	S. Res	M	18	2004	45	18.0	4.50E+01
Krahn et al. 2007	L78	S. Res	M	15	2004	45	15.2	2.20E+01
Krahn et al. 2007	L85	S. Res	M	15	2006	45	24.8	5.00E+01
Present Study	J37	S. Res	F	15	2015	209	64.3	3.01E+00
Present Study	L103	S. Res	F	13	2015	209	64.3	1.33E+01
Present Study	Blubber	S. Res	F	18	2015	209	64.3	4.41E+01
Present Study	K 22	S. Res	F	29	2015	209	64.3	1.42E+01
Present Study	L 116	S. Res	M	6	2015	209	64.3	4.75E+01
Present Study	K 13	S. Res	F	44	2015	209	64.3	4.83E+00
Present Study	K 25	S. Res	M	25	2015	209	64.3	1.03E+01
Present Study	J 49	S. Res	M	4	2015	209	64.3	2.77E+01
Present Study	L 72	S. Res	F	30	2015	209	64.3	9.66E+00
Ross et al. 2013	T14	Bigg's	M	29	1993	205	64.3	2.85E+02
Ross et al. 2013	T138	Bigg's	M	26	1993	205	64.3	1.76E+02

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg\cdot kg^{-1}{}_{lw}$)
Ross et al. 2013	T2B	Bigg's	F	14	1993	205	64.3	2.62E+02
Ross et al. 2013	T44	Bigg's	M	18	1996	205	64.3	8.77E+01
Ross et al. 2013	T54	Bigg's	M	24	1996	205	64.3	2.15E+02
Ross et al. 2013	T142	Bigg's	M	29	1996	205	64.3	1.79E+02
Ross et al. 2013	T2	Bigg's	F	46	1996	205	64.3	4.76E+01
Ross et al. 2013	T69	Bigg's	F	22	1996	205	64.3	1.85E+01
Ross et al. 2013	T28	Bigg's	F	24	1996	205	64.3	5.27E+01
Ross et al. 2013	T60A	Bigg's	*	*	1996	205	64.3	7.01E+00
Ross et al. 2013	T12A	Bigg's	M	14	1996	205	64.3	3.34E+00
Ross et al. 2013	T140	Bigg's	F	22	1996	205	64.3	5.81E+00
Ross et al. 2013	T55	Bigg's	F	22	1996	205	64.3	1.16E+02
Ross et al. 2013	T29	Bigg's	M	40	1996	205	64.3	2.52E+02
Ross et al. 2013	T162	Bigg's	M	31	2002	205	64.3	6.59E+01
Ross et al. 2013	T030B	Bigg's	F	6	2002	205	64.3	6.65E+01
Ross et al. 2013	T030	Bigg's	F	35	2002	205	64.3	3.81E+01
Ross et al. 2013	T109B	Bigg's	F	6	2002	205	64.3	7.67E+01
Ross et al. 2013	T059A	Bigg's	F	7	2002	205	64.3	8.91E+01
Ross et al. 2013	T101A	Bigg's	M	10	2003	205	64.3	3.84E+01
Ross et al. 2013	T021	Bigg's	F	35	2003	205	64.3	2.89E+02
Ross et al. 2013	T74	Bigg's	M	24	2003	205	64.3	6.92E+01
Ross et al. 2013	T007	Bigg's	F	43	2004	205	64.3	1.45E+02
Ross et al. 2013	T188	Bigg's	F	15	2004	205	64.3	1.11E+02
Ross et al. 2013	T141	Bigg's	F	15	2004	205	64.3	1.06E+02
Ross et al. 2013	TO23	Bigg's	F	42	2007	205	64.3	5.70E+01
Ross et al. 2013	T055A	Bigg's	M	18	2007	205	64.3	4.43E+01
Ross et al. 2013	TO69E	Bigg's	*	3	2007	205	64.3	7.52E+01
Ross et al. 2013	TO69C	Bigg's	M	12	2007	205	64.3	9.51E+01
Ross et al. 2013	T150	Bigg's	M	18	2008	205	64.3	8.81E+01
Ross et al. 2013	T060D	Bigg's	M	4	2008	205	64.3	2.14E+02

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg \cdot kg^{-1}{}_{lw}$)
Ross et al. 2013	T060C	Bigg's	*	7	2008	205	64.3	1.21E+02
Ross et al. 2013	T049A	Bigg's	M	22	2008	205	64.3	5.48E+01
Ross et al. 2013	T087	Bigg's	M	45	2008	205	64.3	2.50E+02
Ross et al. 2013	T055B	Bigg's	F	14	2008	205	64.3	2.32E+02
Ross et al. 2013	T055C	Bigg's	*	4	2008	205	64.3	1.81E+02
Ross et al. 2013	T090	Bigg's	F	18	2008	205	64.3	6.94E+01

*Unknown

Table E.2 Concentrations of total polybrominated diphenyl ethers (ΣPBDE) measured in the blubber of killer whales from 1993 to 2015.

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPBDE ($mg \cdot kg^{-1}{}_{lw}$)
Rayne et al. 2004	*	N. Res	F	5	1993	37	48.0	2.83E+00
Rayne et al. 2004	*	N. Res	F	10	1993	37	48.0	7.35E-02
Rayne et al. 2004	*	N. Res	F	12	1993	37	48.0	1.20E-01
Rayne et al. 2004	*	N. Res	F	38	1993	37	45.9	2.58E-02
Rayne et al. 2004	*	N. Res	F	43	1993	37	48.0	2.34E-02
Rayne et al. 2004	*	N. Res	F	56	1993	37	59.1	9.32E-02
Rayne et al. 2004	*	N. Res	F	57	1993	37	48.0	6.29E-02
Rayne et al. 2004	*	N. Res	F	69	1993	37	48.0	9.10E-02
Rayne et al. 2004	*	N. Res	M	1	1993	37	22.9	1.05E-01
Rayne et al. 2004	*	N. Res	M	5	1993	37	48.0	2.27E-01
Rayne et al. 2004	*	N. Res	M	13	1993	37	60.5	1.11E-01
Rayne et al. 2004	*	N. Res	M	19	1993	37	67.9	4.97E-02
Rayne et al. 2004	*	N. Res	M	24	1993	37	65.0	3.73E-02
Rayne et al. 2004	*	N. Res	M	28	1993	37	48.0	9.56E-02
Rayne et al. 2004	*	N. Res	M	36	1993	37	72.4	3.14E-01
Rayne et al. 2004	*	N. Res	M	38	1993	37	76.3	1.03E-01
Rayne et al. 2004	*	N. Res	M	38	1993	37	72.3	1.26E-01

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPBDE ($mg \cdot kg^{-1}_{lw}$)
Rayne et al. 2004	*	N. Res	M	38	1993	37	75.4	7.58E-01
Rayne et al. 2004	*	N. Res	M	40	1993	37	81.9	2.68E-02
Rayne et al. 2004	*	N. Res	M	7	1994	37	48.0	5.14E-01
Rayne et al. 2004	*	N. Res	M	10	1994	37	41.5	1.75E-01
Ross et al. 2013	A62	N. Res	F	10	2003	78	64.3	3.43E-01
Ross et al. 2013	A55	N. Res	M	13	2003	78	64.3	1.24E-01
Ross et al. 2013	A60	N. Res	M	11	2003	78	64.3	1.47E+00
Ross et al. 2013	I68	N. Res	F	11	2003	78	64.3	7.24E-01
Ross et al. 2013	A33	N. Res	M	32	2003	78	64.3	3.03E-01
Ross et al. 2013	I67	N. Res	M	12	2003	78	64.3	7.67E-01
Ross et al. 2013	I42	N. Res	M	20	2003	78	64.3	4.84E-01
Ross et al. 2013	A74	N. Res	M	3	2003	78	64.3	8.53E-01
Ross et al. 2013	R28	N. Res	M	15	2003	78	64.3	2.37E-01
Ross et al. 2013	C16	N. Res	F	15	2004	78	64.3	4.36E-01
Ross et al. 2013	A71	N. Res	M	5	2004	78	64.3	4.91E-01
Ross et al. 2013	B14	N. Res	F	16	2007	78	64.3	4.54E-01
Ross et al. 2013	A71	N. Res	M	8	2007	78	64.3	1.71E+00
Ross et al. 2013	A61	N. Res	M	13	2007	78	64.3	1.40E+00
Ross et al. 2013	C22	N. Res	M	10	2007	78	64.3	1.52E+00
Ross et al. 2013	I64	N. Res	M	17	2007	78	64.3	7.64E-01
Ross et al. 2013	I80	N. Res	F	10	2007	78	64.3	2.45E+00
Ross et al. 2013	I46	N. Res	M	22	2007	78	64.3	3.26E-01
Ross et al. 2013	R31	N. Res	M	10	2007	78	64.3	7.76E-01
Ross et al. 2013	R30	N. Res	M	13	2007	78	64.3	4.62E-01
Ross et al. 2013	R43	N. Res	M	5	2007	78	64.3	1.35E+00
Ross et al. 2013	I35	N. Res	F	33	2007	78	64.3	4.90E-01
Ross et al. 2013	I110	N. Res	**	2	2007	78	64.3	1.21E+00
Rayne et al. 2004	*	S. Res	M	37	1993	37	48.0	3.11E-01

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPBDE ($mg \cdot kg^{-1}_{lw}$)
Rayne et al. 2004	*	S. Res	M	39	1993	37	48.0	1.25E+00
Rayne et al. 2004	*	S. Res	M	13	1995	37	48.0	2.42E-01
Rayne et al. 2004	*	S. Res	M	18	1995	37	48.0	1.10E+00
Rayne et al. 2004	*	S. Res	M	44	1995	37	48.0	1.81E+00
Ross et al. 2013	L78	S. Res	M	15	2004	78	64.3	1.06E+00
Ross et al. 2013	L74	S. Res	M	18	2004	78	64.3	1.41E+00
Ross et al. 2013	L71	S. Res	M	18	2004	78	64.3	1.29E+00
Krahn et al. 2009	L71	S. Res	M	18	2004	10	9.60	2.60E+00
Krahn et al. 2009	L74	S. Res	M	18	2004	10	18.0	3.10E+00
Krahn et al. 2009	L78	S. Res	M	15	2004	10	15.2	2.60E+00
Krahn et al. 2009	J19	S. Res	F	27	2006	10	29.4	7.50E+00
Krahn et al. 2009	J39	S. Res	M	3	2006	10	40.9	1.50E+01
Krahn et al. 2009	J1	S. Res	M	55	2006	10	21.9	6.80E+00
Krahn et al. 2009	J27	S. Res	M	15	2006	10	30.4	6.30E+00
Krahn et al. 2009	L57	S. Res	M	29	2006	10	19.4	3.30E+00
Krahn et al. 2009	L85	S. Res	M	15	2006	10	24.8	2.50E+00
Present Study	L103	S. Res	F	13	2015	46	64.3	1.75E+00
Present Study	Blubber	S. Res	F	18	2015	46	64.3	4.59E+00
Present Study	K22	S. Res	F	29	2015	46	64.3	1.81E+00
Present Study	L116	S. Res	F	6	2015	46	64.3	4.40E+00
Present Study	K13	S. Res	F	44	2015	46	64.3	1.06E+00
Present Study	J49	S. Res	F	4	2015	46	64.3	6.62E+00
Present Study	J37	S. Res	M	15	2015	46	64.3	4.93E-01
Present Study	K25	S. Res	M	25	2015	46	64.3	1.57E+00
Present Study	L72	S. Res	M	30	2015	46	64.3	7.44E-01
Rayne et al. 2004	*	Bigg’s	F	11	1993	37	48.0	5.07E-01
Rayne et al. 2004	*	Bigg’s	M	25	1993	37	35.2	2.25E+00
Rayne et al. 2004	*	Bigg’s	F	14	1994	37	38.0	6.23E-01

Source	Animal ID	Ecotype	Sex	Age	Year	Number of Congeners Analyzed	Lipid %	ΣPBDE ($mg \cdot kg^{-1}_{lw}$)
Rayne et al. 2004	*	Bigg's	F	15	1994	37	38.2	2.90E+00
Rayne et al. 2004	*	Bigg's	F	20	1994	37	48.0	1.62E-01
Rayne et al. 2004	*	Bigg's	M	22	1994	37	46.3	2.83E-01
Rayne et al. 2004	*	Bigg's	M	27	1994	37	48.0	4.92E-01
Rayne et al. 2004	*	Bigg's	F	1	1996	37	13.2	1.22E+00
Rayne et al. 2004	*	Bigg's	F	22	1996	37	20.6	2.39E-01
Rayne et al. 2004	*	Bigg's	M	14	1996	37	40.9	9.14E-01
Rayne et al. 2004	*	Bigg's	M	18	1996	37	4.40	5.70E-01
Rayne et al. 2004	*	Bigg's	M	40	1996	37	48.0	1.58E+00
Rayne et al. 2004	*	Bigg's	F	1	1997	37	39.1	5.42E-01
Ross et al. 2013	T101A	Bigg's	M	10	2003	78	64.3	1.60E+00
Ross et al. 2013	T021	Bigg's	F	35	2003	78	64.3	1.73E+01
Ross et al. 2013	T74	Bigg's	M	24	2003	78	64.3	1.68E+00
Ross et al. 2013	T007	Bigg's	F	43	2004	78	64.3	4.72E+00
Ross et al. 2013	T188	Bigg's	F	15	2004	78	64.3	1.73E+00
Ross et al. 2013	T141	Bigg's	F	15	2004	78	64.3	3.18E+00
Ross et al. 2013	TO23	Bigg's	F	42	2007	78	64.3	3.57E+00
Ross et al. 2013	T055A	Bigg's	M	18	2007	78	64.3	1.40E+00
Ross et al. 2013	TO69E	Bigg's	**	3	2007	78	64.3	8.97E+00
Ross et al. 2013	TO69C	Bigg's	M	12	2007	78	64.3	5.49E+00

* Information not given in publication, ** Unknown

Table E.3 Concentrations of total isomers of hexabromocyclododecane (ΣHBCDD) measured in the blubber of killer whales for the present study.

Source	Animal ID	Ecotype	Sex	Age	Year	Lipid %	ΣHBCDD ($mg \cdot kg^{-1}_{lw}$)
Present Study	Blubber	S. Res	F	18	2015	64.3	6.84E-02
Present Study	K22	S. Res	F	29	2015	64.3	6.62E-02
Present Study	J37	S. Res	F	15	2015	64.3	5.14E-02

Present Study	L103	S. Res	F	13	2015	64.3	6.91E-02
Present Study	L116	S. Res	M	6	2015	64.3	1.20E-01
Present Study	K13	S. Res	F	44	2015	64.3	4.22E-02
Present Study	K25	S. Res	M	25	2015	64.3	5.57E-02
Present Study	J49	S. Res	M	4	2015	64.3	1.17E-01
Present Study	L72	S. Res	F	30	2015	64.3	5.73E-02

Table E.4 **Concentrations of Dechlorane, Dechlorane Plus, Dechlorane 602 (Dec 602), 1,2-dibromobenzene (1,2-DiBB), pentabromotoluene (PBT) and tetrabromo-o-chlorotoluene (TBCT) measured in killer whale blubber for the present study.**

Source	Animal ID	Ecotype	Sex	Age	Year	Lipid %	Dechlorane ($mg \cdot kg^{-1}_{lw}$)	Dechlorane Plus ($mg \cdot kg^{-1}_{lw}$)	Dec 602 ($mg \cdot kg^{-1}_{lw}$)	1,2-DiBB ($mg \cdot kg^{-1}_{lw}$)	PBT ($mg \cdot kg^{-1}_{lw}$)	TBCT ($mg \cdot kg^{-1}_{lw}$)
Present Study	Blubber	S. Res	F	18	2015	64.3	9.98E-02	9.80E-04	5.58E-03	3.73E-03	6.22E-04	1.38E-03
Present Study	K22	S. Res	F	29	2015	64.3	1.08E-01	6.67E-03	5.10E-03	7.82E-02	1.23E-02	1.07E-02
Present Study	J37	S. Res	F	15	2015	64.3	3.17E-02	6.78E-03	2.75E-03	7.96E-02	1.01E-02	3.96E-03
Present Study	L103	S. Res	F	13	2015	64.3	7.20E-02	7.14E-03	2.38E-03	7.48E-02	9.88E-03	1.06E-02
Present Study	L116	S. Res	M	6	2015	64.3	1.85E-01	1.10E-02	4.65E-03	9.92E-02	9.88E-03	2.05E-02
Present Study	K13	S. Res	F	44	2015	64.3	3.48E-02	1.21E-02	2.32E-03	7.67E-02	9.35E-03	4.96E-03
Present Study	K25	S. Res	M	25	2015	64.3	4.88E-02	8.02E-03	2.80E-03	9.32E-02	9.10E-03	1.01E-02
Present Study	J49	S. Res	M	4	2015	64.3	5.16E-02	9.20E-03	1.22E-03	9.42E-02	1.91E-02	1.09E-02
Present Study	L72	S. Res	F	30	2015	64.3	6.67E-02	7.42E-03	3.48E-03	9.44E-02	1.31E-02	5.24E-03

Table E.5 Concentrations of total polychlorinated biphenyls (ΣPCB) measured in Chinook salmon samples from 2000 to 2014.

Source	Year	Number of Congeners Analyzed	Lipid %	ΣPCB ($mg \cdot kg^{-1}{}_{lw}$)
Cullon et al. 2009	2000	209	15.1	6.48E-02
Cullon et al. 2009	2000	209	5.14	9.23E-02
Cullon et al. 2009	2000	209	11.9	5.68E-02
Cullon et al. 2009	2000	209	7.93	2.17E-01
Cullon et al. 2009	2000	209	11.9	1.11E-01
Cullon et al. 2009	2000	209	9.76	7.44E-02
Cullon et al. 2009	2000	209	2.69	1.27E+00
Cullon et al. 2009	2000	209	8.80	5.22E-01
Cullon et al. 2009	2000	209	6.44	5.90E-01
Cullon et al. 2009	2000	209	3.29	2.46E+00
Cullon et al. 2009	2000	209	4.72	4.62E-01
Cullon et al. 2009	2000	209	6.26	8.58E-01
Present Study	2014	209	7.25	3.03E-01
Present Study	2014	209	6.55	2.84E-01
Present Study	2014	209	2.08	7.27E-01
Present Study	2014	209	8.55	2.47E-01
Present Study	2014	209	8.72	1.55E-01
Present Study	2014	209	9.20	2.25E-01
Present Study	2014	209	11.9	2.80E-01

Table E.6 Concentrations of total polybrominated diphenyl ethers (ΣPBDE) measured in Chinook salmon samples from 2002 to 2014.

Source	Year	Number of Congeners Analyzed	Lipid %	ΣPBDE ($mg \cdot kg^{-1}{}_{lw}$)
Unpublished Axys Data	2002	44	6.20	2.85E-01
Present Study	2014	46	7.25	8.14E-02
Present Study	2014	46	6.55	7.05E-02

Present Study	2014	46	2.08	2.49E-01
Present Study	2014	46	8.55	4.84E-02
Present Study	2014	46	8.72	2.94E-02
Present Study	2014	46	9.20	4.95E-02
Present Study	2014	46	11.9	5.09E-02

Table E.7 Concentrations of total isomers of hexabromocyclododecane (ΣHBCDD) measured in Chinook salmon samples for the present study.

Source	Year	Lipid %	ΣHBCDD ($mg{\cdot}kg^{-1}{}_{lw}$)
Present Study	2014	7.25	6.99E-03
Present Study	2014	6.55	7.66E-03
Present Study	2014	2.08	2.00E-02
Present Study	2014	8.55	6.93E-03
Present Study	2014	8.72	5.74E-03
Present Study	2014	9.20	6.05E-03
Present Study	2014	11.9	4.86E-03

Note: Dechlorane, Dechlorane Plus, Dec 602, 1,2-DiBB, PBT and TBCT were not detected in any of the Chinook salmon samples for the present study

Appendix F.

Linear Regression Analyses for Reproductive and Post-Reproductive Female Killer Whales

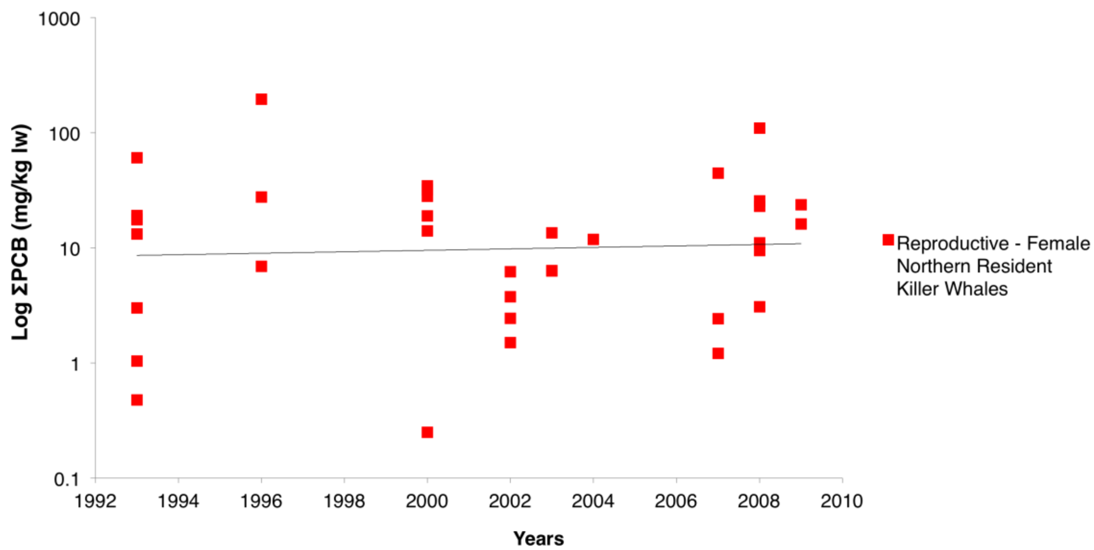

Figure F.1 **Log concentrations (mg·kg^{-1} lipid weight [lw]) of total polychlorinated biphenyls (ΣPCB) in reproductive female northern resident killer whales plotted over time from 1993 to 2015 (◆). The linear regression line shows the linear relationship between the log-transformed concentrations of ΣPCB and time.**

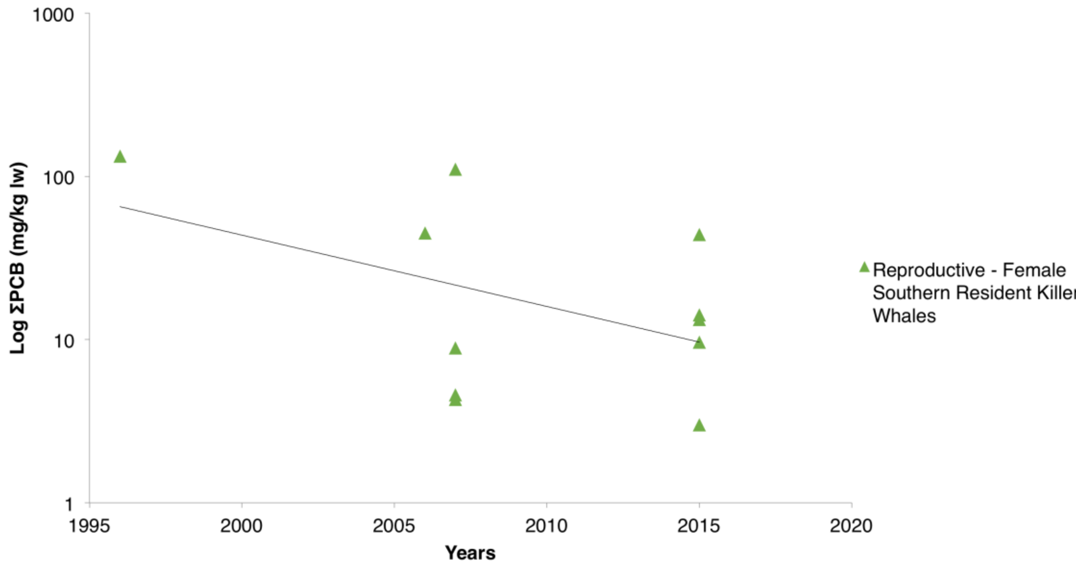

Figure F.2 **Log concentrations (mg·kg^{-1} lipid weight [lw]) of total polychlorinated biphenyls (ΣPCB) in reproductive female southern resident killer whales plotted over time from 1993 to 2015 (◆). The linear regression line shows the linear relationship between the log-transformed concentrations of ΣPCB and time.**

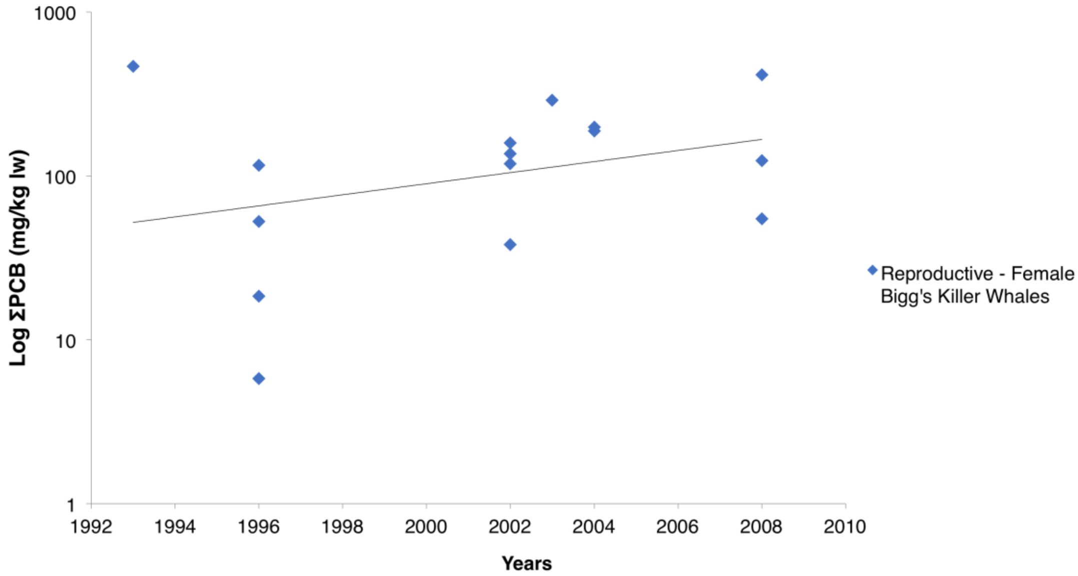

Figure F.3 **Log concentrations (mg·kg^{-1} lipid weight [lw]) of total polychlorinated biphenyls (ΣPCB) in reproductive female Bigg's killer whales plotted over time from 1993 to 2015 (◆). The linear regression line shows the linear relationship between the log-transformed concentrations of ΣPCB and time.**

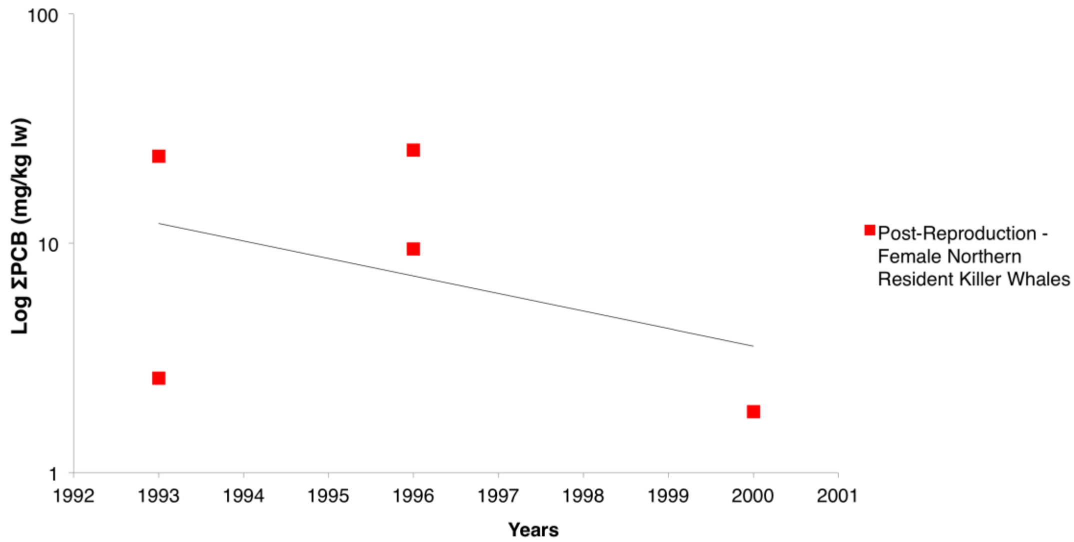

Figure F.4 **Log concentrations (mg·kg^{-1} lipid weight [lw]) of total polychlorinated biphenyls (ΣPCB) in post-reproductive female northern resident killer whales plotted over time from 1993 to 2015 (◆). The linear regression line shows the linear relationship between the log-transformed concentrations of ΣPCB and time.**

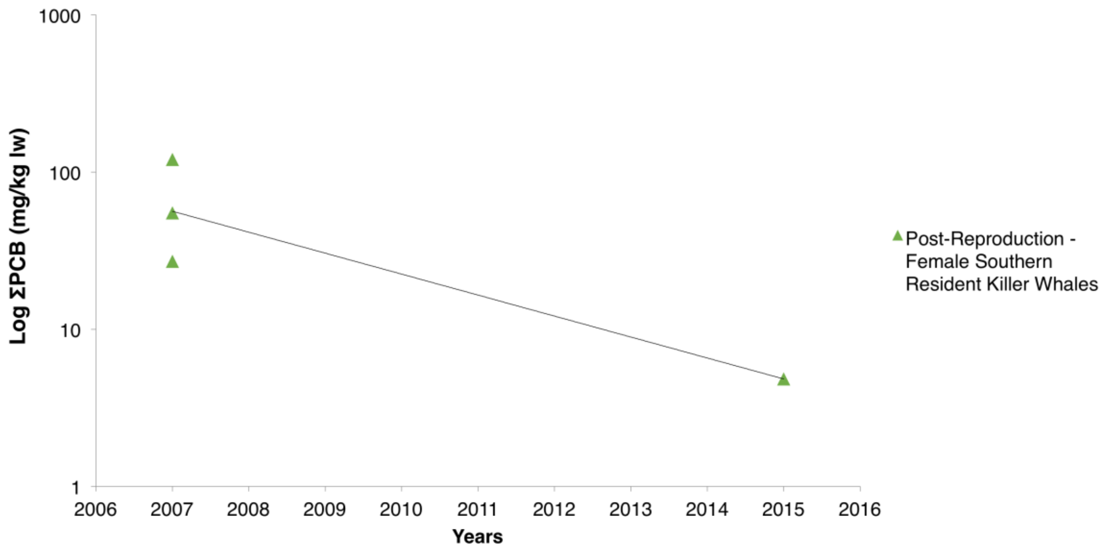

Figure F.5 **Log concentrations (mg·kg^{-1} lipid weight [lw]) of total polychlorinated biphenyls (ΣPCB) in post-reproductive female southern resident killer whales plotted over time from 1993 to 2015 (◆). The linear regression line shows the linear relationship between the log-transformed concentrations of ΣPCB and time.**

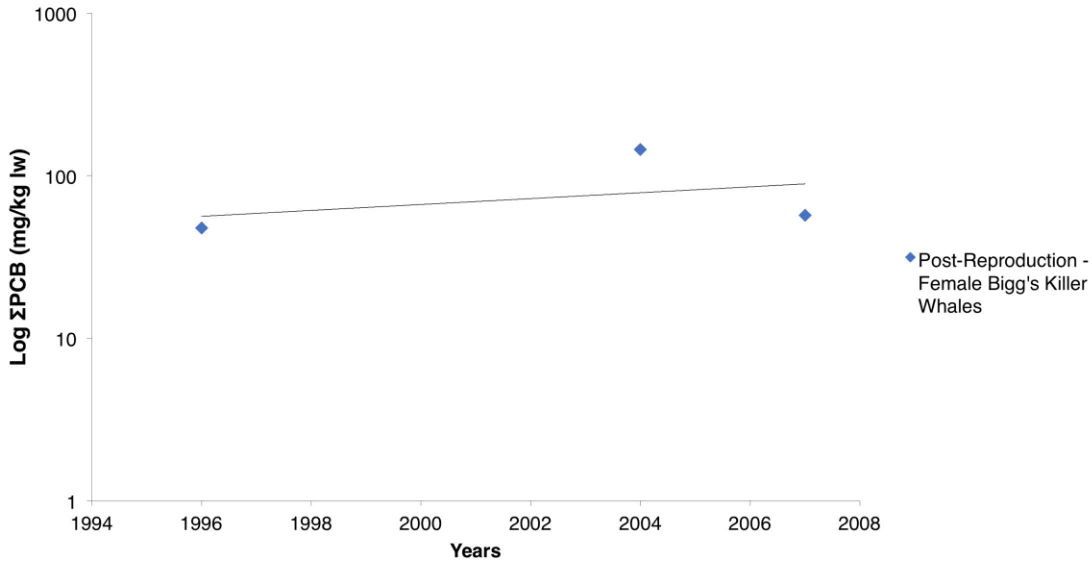

Figure F.6 **Log concentrations (mg·kg^{-1} lipid weight [lw]) of total polychlorinated biphenyls (ΣPCB) in post-reproductive female Bigg's killer whales plotted over time from 1993 to 2015 (◆). The linear regression line shows the linear relationship between the log-transformed concentrations of ΣPCB and time.**

Appendix G.

Mean Results of One-Way ANOVA Tests for Killer Whales

Table G.1 **Mean results of one-way ANOVA tests comparing the mean age of killer whales over the sampled years. The results have been split by sex and ecotype, and are based on the data set regarding polychlorinated biphenyls (PCBs) found in killer whales. The mean age for each year, the number of individual killer whales sampled each year, and the related standard errors and 95% confidence limits are given. NR = northern resident, SR = southern resident.**

Ecotype	Sex	Level	Number	Mean Age (Years)	Standard Error	Lower 95%	Upper 95%
NR	Female	1993	10	2.14E+01	5.06E+00	1.11E+01	3.17E+01
NR	Female	1996	5	3.48E+01	7.16E+00	2.02E+01	4.94E+01
NR	Female	2000	7	1.26E+01	6.05E+00	2.60E-01	2.49E+01
NR	Female	2002	4	2.03E+01	8.00E+00	3.97E+00	3.65E+01
NR	Female	2003	3	1.73E+01	9.24E+00	-1.47E+00	3.61E+01
NR	Female	2004	1	1.50E+01	1.60E+01	-1.76E+01	4.76E+01
NR	Female	2007	3	1.97E+01	9.24E+00	8.70E-01	3.85E+01
NR	Female	2008	6	1.25E+01	6.53E+00	-7.90E-01	2.58E+01
NR	Female	2009	2	1.25E+01	9.24E+00	-7.47E+00	3.01E+01
SR	Female	1996	2	2.85E+01	1.66E+01	-7.70E+00	6.47E+01
SR	Female	2006	1	2.70E+01	2.35E+01	-2.42E+01	7.82E+01
SR	Female	2007	7	4.11E+01	8.88E+00	2.18E+01	6.05E+01
SR	Female	2015	6	2.48E+01	9.59E+00	3.93E+00	4.57E+01
Bigg's	Female	1993	1	1.40E+01	1.26E+01	-1.41E+01	4.21E+01
Bigg's	Female	1996	5	2.72E+01	5.64E+00	1.46E+01	3.98E+01
Bigg's	Female	2002	4	1.35E+01	6.31E+00	-5.60E-01	2.76E+01
Bigg's	Female	2003	1	3.50E+01	1.26E+01	6.88E+00	6.31E+01
Bigg's	Female	2004	3	2.43E+01	7.29E+00	8.10E+00	4.06E+01
Bigg's	Female	2007	1	4.20E+01	1.26E+01	1.39E+01	7.01E+01

Ecotype	Sex	Level	Number	Mean Age (Years)	Standard Error	Lower 95%	Upper 95%
Bigg's	Female	2008	2	1.60E+01	8.92E+00	-3.88E+00	3.59E+01
NR	Male	1993	8	2.16E+01	3.75E+00	1.40E+01	2.92E+01
NR	Male	1996	6	2.77E+01	4.33E+00	1.89E+01	3.64E+01
NR	Male	2000	3	6.67E+00	6.12E+00	-5.74E+00	1.91E+01
NR	Male	2002	2	1.35E+01	7.50E+00	-1.69E+00	2.87E+01
NR	Male	2003	7	1.51E+01	4.01E+00	7.02E+00	2.33E+01
NR	Male	2004	1	5.00E+00	1.06E+01	-1.65E+01	2.65E+01
NR	Male	2007	8	1.23E+01	3.75E+00	4.65E+00	1.98E+01
NR	Male	2008	10	1.83E+01	3.35E+00	1.15E+01	2.51E+01
SR	Male	1993	2	3.85E+01	9.14E+00	1.92E+01	5.78E+01
SR	Male	1996	2	3.30E+01	9.14E+00	1.37E+01	5.23E+01
SR	Male	2000	1	2.30E+01	1.29E+01	-4.28E+00	5.03E+01
SR	Male	2004	6	1.70E+01	5.28E+00	5.86E+00	2.81E+01
SR	Male	2006	5	2.34E+01	5.78E+00	1.12E+01	3.56E+01
SR	Male	2007	5	1.62E+01	5.78E+00	4.00E+00	2.84E+01
SR	Male	2015	3	1.17E+01	7.46E+00	-4.08E+00	2.74E+01
?	Male	1993	2	2.75E+01	8.37E+00	8.85E+00	4.62E+01
Bigg's	Male	1996	5	2.50E+01	5.29E+00	1.32E+01	3.68E+01
Bigg's	Male	2002	1	3.10E+01	1.18E+01	4.62E+00	5.74E+01
Bigg's	Male	2003	2	1.70E+01	8.37E+00	-1.65E+00	3.57E+01
Bigg's	Male	2007	2	1.50E+01	8.37E+00	-3.65E+00	3.37E+01
Bigg's	Male	2008	4	2.23E+01	5.92E+00	9.06E+00	3.54E+01

Table G.2 **Mean results of one-way ANOVA tests comparing the mean age of killer whales over the sampled years. The results have been split by sex and ecotype, and are based on the data set regarding polybrominated diphenyl ethers (PBDEs) found in killer whales. The mean age for each year, the number of individual killer whales sampled each year, and the related standard errors and 95% confidence limits are given. NR = northern resident, SR = southern resident.**

Ecotype	Sex	Level	Number	Mean Age (Years)	Standard Error	Lower 95%	Upper 95%
NR	Female	1993	8	3.63E+01	7.48E+00	1.96E+01	5.29E+01
NR	Female	2003	2	1.05E+01	1.50E+01	-2.29E+01	4.39E+01
NR	Female	2004	1	1.50E+01	2.12E+01	-3.22E+01	6.22E+01
NR	Female	2007	3	1.97E+01	1.22E+01	-7.56E+00	4.69E+01
Bigg's	Female	1993	1	1.10E+01	1.24E+01	-2.08E+01	4.28E+01
Bigg's	Female	1994	3	1.63E+01	7.14E+00	-2.01E+00	3.47E+01
Bigg's	Female	1996	2	1.15E+01	8.74E+00	-1.10E+01	3.40E+01
Bigg's	Female	1997	1	1.00E+00	1.24E+01	-3.08E+01	3.28E+01
Bigg's	Female	2003	1	3.50E+01	1.24E+01	3.23E+00	6.68E+01
Bigg's	Female	2004	3	2.43E+01	7.14E+00	5.99E+00	4.27E+01
Bigg's	Female	2007	1	4.20E+01	1.24E+01	1.02E+01	7.38E+01
NR	Male	1993	11	2.55E+01	3.24E+00	1.88E+01	3.21E+01
NR	Male	1994	2	8.50E+00	7.59E+00	-7.19E+00	2.42E+01
NR	Male	2003	7	1.51E+01	4.06E+00	6.75E+00	2.35E+01
NR	Male	2004	1	5.00E+00	1.07E+01	-1.72E+01	2.72E+01
NR	Male	2007	7	1.33E+01	4.06E+00	4.90E+00	2.17E+01
SR	Male	1993	2	3.80E+01	9.01E+00	1.87E+01	5.73E+01
SR	Male	1995	3	2.50E+01	7.35E+00	9.23E+00	4.08E+01
SR	Male	2004	6	1.70E+01	5.20E+00	5.85E+00	2.82E+01
SR	Male	2006	5	2.34E+01	5.70E+00	1.12E+01	3.56E+01
SR	Male	2015	3	2.33E+01	7.35E+00	7.56E+00	3.91E+01
Bigg's	Male	1993	1	2.50E+01	1.02E+01	-1.23E+00	5.12E+01
Bigg's	Male	1994	2	2.45E+01	7.22E+00	5.95E+00	4.30E+01
Bigg's	Male	1996	3	2.40E+01	5.89E+00	8.86E+00	3.91E+01

Bigg's	Male	2003	2	1.70E+01	7.22E+00	-1.55E+00	3.55E+01
Bigg's	Male	2007	2	1.50E+01	7.22E+00	-3.55E+00	3.35E+01

Appendix H.

Individual Ratio Results

Table H.1 **The ratio of total polybrominated diphenyl ethers to total polychlorinated biphenyls (ΣPBDEs/ΣPCBs) in each individual whale that was sampled for both ΣPBDEs and ΣPCBs in the same year. Year of collection, killer whale ID, sex of whale and ecotype are shown for each sample. NR = northern resident, SR = southern resident.**

ΣPBDEs /ΣPCBs	Year	Animal ID	Sex	Ecotype
2.70E-02	2003	A33	M	NR
4.78E-02	2003	A55	M	NR
9.66E-02	2003	A60	M	NR
9.69E-02	2003	A62	F	NR
1.25E-01	2003	A74	M	NR
6.77E-02	2003	I42	M	NR
3.24E-02	2003	I67	M	NR
9.57E-02	2003	I68	F	NR
6.80E-02	2003	R28	M	NR
5.96E-02	2003	T021	F	Bigg's
4.17E-02	2003	T101A	M	Bigg's
2.43E-02	2003	T74	M	Bigg's
1.50E-01	2004	A71	M	NR
6.56E-02	2004	C16	F	NR
7.24E-02	2004	L71	M	SR
6.32E-02	2004	L74	M	SR
1.25E-01	2004	L78	M	SR
3.25E-02	2004	T007	F	Bigg's
3.01E-02	2004	T141	F	Bigg's
1.56E-02	2004	T188	F	Bigg's
3.78E-02	2006	J1	M	SR

ΣPBDEs /ΣPCBs	Year	Animal ID	Sex	Ecotype
1.67E-01	2006	J19	F	SR
8.51E-02	2006	J27	M	SR
4.41E-01	2006	J39	M	SR
5.89E-02	2006	L57	M	SR
5.00E-02	2006	L85	M	SR
1.44E-01	2007	A61	M	NR
2.09E-01	2007	A71	M	NR
3.32E-01	2007	B14	F	NR
1.78E-01	2007	C22	M	NR
2.36E-01	2007	I110	Unknown	NR
4.05E-01	2007	I35	F	NR
4.52E-01	2007	I46	M	NR
1.12E-01	2007	I64	M	NR
9.81E-02	2007	I80	F	NR
7.01E-02	2007	R30	M	NR
3.65E-02	2007	R31	M	NR
1.17E-01	2007	R43	Unknown	NR
6.25E-02	2007	TO23	F	Bigg's
5.77E-02	2007	TO69C	M	Bigg's
1.19E-01	2007	TO69E	Unknown	Bigg's
1.64E-01	2015	J37	M	SR
2.39E-01	2015	J49	F	SR
2.20E-01	2015	K13	F	SR
1.27E-01	2015	K22	F	SR
1.53E-01	2015	K25	M	SR
7.70E-02	2015	L 72	F	SR
1.31E-01	2015	L103	F	SR
9.26E-02	2015	L116	F	SR

Table H.2 **Concentrations of total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs) in each individual female killer whale divided by the corresponding toxicity reference value (TRV) for each contaminant. Individual killer whale ID, ecotype, sex and year of collection are given for each sample. The average TRV of 1.5 mg·kg^{-1} lipid weight for both ΣPCBs and ΣPBDEs was used for calculations. NR = northern resident, SR = southern resident.**

Animal ID	Ecotype	Sex	Year	Concentration/TRV	Contaminant
J20	SR	F	1996	4.87E+01	ΣPCBs
J11	SR	F	1996	2.27E+01	ΣPCBs
J19	SR	F	2006	2.93E+01	ΣPCBs
J22	SR	F	2007	3.00E+00	ΣPCBs
K7	SR	F	2007	7.83E+01	ΣPCBs
K13	SR	F	2007	5.80E+00	ΣPCBs
K36	SR	F	2007	4.04E+01	ΣPCBs
L21	SR	F	2007	3.59E+01	ΣPCBs
L26	SR	F	2007	1.76E+01	ΣPCBs
L67	SR	F	2007	2.80E+00	ΣPCBs
J37	SR	F	2015	1.96E+00	ΣPCBs
L103	SR	F	2015	8.69E+00	ΣPCBs
Blubber	SR	F	2015	2.88E+01	ΣPCBs
K 22	SR	F	2015	9.27E+00	ΣPCBs
K 13	SR	F	2015	3.15E+00	ΣPCBs
L 72	SR	F	2015	6.30E+00	ΣPCBs
A43	NR	F	1993	4.83E+00	ΣPCBs
A23	NR	F	1993	1.68E+00	ΣPCBs
A24	NR	F	1993	3.12E-01	ΣPCBs
A54	NR	F	1993	6.50E+00	ΣPCBs
A11	NR	F	1993	6.80E-01	ΣPCBs
A35	NR	F	1993	1.10E+00	ΣPCBs
A48	NR	F	1993	6.96E+00	ΣPCBs
A52	NR	F	1993	2.21E+01	ΣPCBs
A56	NR	F	1993	6.39E+00	ΣPCBs

Animal ID	Ecotype	Sex	Year	Concentration/TRV	Contaminant
A9	NR	F	1993	1.57E+01	ΣPCBs
I2	NR	F	1996	6.16E+00	ΣPCBs
C5	NR	F	1996	1.66E+01	ΣPCBs
C10	NR	F	1996	4.50E+00	ΣPCBs
A42	NR	F	1996	1.01E+01	ΣPCBs
A57	NR	F	1996	7.13E+01	ΣPCBs
A70	NR	F	2000	2.68E+00	ΣPCBs
A69	NR	F	2000	6.91E+00	ΣPCBs
I51	NR	F	2000	5.12E+00	ΣPCBs
G51	NR	F	2000	1.02E+01	ΣPCBs
I80	NR	F	2000	1.26E+01	ΣPCBs
I15	NR	F	2000	1.21E+00	ΣPCBs
I63	NR	F	2000	1.17E+01	ΣPCBs
D12	NR	F	2002	9.84E-01	ΣPCBs
I 21	NR	F	2002	1.59E+00	ΣPCBs
D13	NR	F	2002	2.27E+00	ΣPCBs
I 50	NR	F	2002	2.45E+00	ΣPCBs
D 09	NR	F	2003	4.15E+00	ΣPCBs
I68	NR	F	2003	4.94E+00	ΣPCBs
A62	NR	F	2003	2.31E+00	ΣPCBs
C16	NR	F	2004	4.33E+00	ΣPCBs
B14	NR	F	2007	8.93E-01	ΣPCBs
I80	NR	F	2007	1.63E+01	ΣPCBs
I35	NR	F	2007	7.89E-01	ΣPCBs
A67	NR	F	2008	3.48E+00	ΣPCBs
A51	NR	F	2008	7.24E+00	ΣPCBs
C24	NR	F	2008	4.00E+01	ΣPCBs
A72	NR	F	2008	9.45E+00	ΣPCBs
A75	NR	F	2008	8.41E+00	ΣPCBs
A54	NR	F	2008	1.15E+00	ΣPCBs

Animal ID	Ecotype	Sex	Year	Concentration/TRV	Contaminant
R39	NR	F	2009	5.14E+00	ΣPCBs
R35	NR	F	2009	8.65E+00	ΣPCBs
R29	NR	F	2009	5.87E+00	ΣPCBs
T2B	Bigg's	F	1993	1.71E+02	ΣPCBs
T2	Bigg's	F	1996	3.10E+01	ΣPCBs
T69	Bigg's	F	1996	1.21E+01	ΣPCBs
T28	Bigg's	F	1996	3.44E+01	ΣPCBs
T140	Bigg's	F	1996	3.79E+00	ΣPCBs
T55	Bigg's	F	1996	7.59E+01	ΣPCBs
T030B	Bigg's	F	2002	4.33E+01	ΣPCBs
T030	Bigg's	F	2002	2.49E+01	ΣPCBs
T109B	Bigg's	F	2002	5.00E+01	ΣPCBs
T059A	Bigg's	F	2002	5.81E+01	ΣPCBs
T021	Bigg's	F	2003	1.89E+02	ΣPCBs
T007	Bigg's	F	2004	9.47E+01	ΣPCBs
T188	Bigg's	F	2004	7.24E+01	ΣPCBs
T141	Bigg's	F	2004	6.89E+01	ΣPCBs
TO23	Bigg's	F	2007	3.72E+01	ΣPCBs
T055B	Bigg's	F	2008	1.51E+02	ΣPCBs
T090	Bigg's	F	2008	4.52E+01	ΣPCBs
Unknown	NR	F	1993	1.89E+00	ΣPBDEs
Unknown	NR	F	1993	4.90E-02	ΣPBDEs
Unknown	NR	F	1993	7.98E-02	ΣPBDEs
Unknown	NR	F	1993	1.72E-02	ΣPBDEs
Unknown	NR	F	1993	1.56E-02	ΣPBDEs
Unknown	NR	F	1993	6.21E-02	ΣPBDEs
Unknown	NR	F	1993	4.19E-02	ΣPBDEs
Unknown	NR	F	1993	6.07E-02	ΣPBDEs
A62	NR	F	2003	2.29E-01	ΣPBDEs
I68	NR	F	2003	4.83E-01	ΣPBDEs

Animal ID	Ecotype	Sex	Year	Concentration/TRV	Contaminant
C16	NR	F	2004	2.91E-01	ΣPBDEs
B14	NR	F	2007	3.02E-01	ΣPBDEs
I80	NR	F	2007	1.63E+00	ΣPBDEs
I35	NR	F	2007	3.27E-01	ΣPBDEs
J19	SR	F	2006	5.00E+00	ΣPBDEs
L103	SR	F	2015	1.16E+00	ΣPBDEs
Blubber	SR	F	2015	3.06E+00	ΣPBDEs
K22	SR	F	2015	1.21E+00	ΣPBDEs
L116	SR	F	2015	2.93E+00	ΣPBDEs
K13	SR	F	2015	7.08E-01	ΣPBDEs
J49	SR	F	2015	4.42E+00	ΣPBDEs
Unknown	Bigg's	F	1993	3.38E-01	ΣPBDEs
Unknown	Bigg's	F	1994	4.15E-01	ΣPBDEs
Unknown	Bigg's	F	1994	1.93E+00	ΣPBDEs
Unknown	Bigg's	F	1994	1.08E-01	ΣPBDEs
Unknown	Bigg's	F	1996	8.16E-01	ΣPBDEs
Unknown	Bigg's	F	1996	1.59E-01	ΣPBDEs
Unknown	Bigg's	F	1997	3.61E-01	ΣPBDEs
T021	Bigg's	F	2003	1.15E+01	ΣPBDEs
T007	Bigg's	F	2004	3.15E+00	ΣPBDEs
T188	Bigg's	F	2004	1.16E+00	ΣPBDEs
T141	Bigg's	F	2004	2.12E+00	ΣPBDEs
TO23	Bigg's	F	2007	2.38E+00	ΣPBDEs

Table H.3 **Concentrations of total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs) in individual male killer whales divided by the corresponding toxicity reference value (TRV) for each contaminant. Individual killer whale ID, ecotype, sex and year of collection are given for each sample. The average TRV of 1.5 mg·kg^{-1} lipid weight for both ΣPCBs and ΣPBDEs was used for calculations. NR = northern resident, SR = southern resident.**

Animal ID	Ecotype	Sex	Year	Contaminant/TRV	Contaminant
A27	NR	M	1993	1.58E+00	ΣPCBs
A60	NR	M	1993	8.57E+00	ΣPCBs
A6	NR	M	1993	1.17E+01	ΣPCBs
B1	NR	M	1993	4.50E+00	ΣPCBs
B2	NR	M	1993	1.76E+01	ΣPCBs
A13	NR	M	1993	1.38E+01	ΣPCBs
A59	NR	M	1993	7.02E+00	ΣPCBs
B12	NR	M	1996	1.30E+01	ΣPCBs
B13	NR	M	1996	1.82E+01	ΣPCBs
A5	NR	M	1996	2.49E+01	ΣPCBs
I5	NR	M	1996	2.46E+01	ΣPCBs
R6	NR	M	1996	3.24E+01	ΣPCBs
H4	NR	M	1996	1.44E+01	ΣPCBs
C17	NR	M	2000	4.50E+00	ΣPCBs
I85	NR	M	2000	3.75E+00	ΣPCBs
C20	NR	M	2000	3.56E+00	ΣPCBs
C18	NR	M	2002	4.09E+00	ΣPCBs
I 52	NR	M	2002	5.37E+00	ΣPCBs
A55	NR	M	2003	1.69E+00	ΣPCBs
A60	NR	M	2003	9.95E+00	ΣPCBs
A33	NR	M	2003	7.32E+00	ΣPCBs
I67	NR	M	2003	1.54E+01	ΣPCBs
I42	NR	M	2003	4.66E+00	ΣPCBs
A74	NR	M	2003	4.46E+00	ΣPCBs
R28	NR	M	2003	2.27E+00	ΣPCBs

Animal ID	Ecotype	Sex	Year	Contaminant/TRV	Contaminant
A71	NR	M	2004	2.13E+00	ΣPCBs
A71	NR	M	2007	5.33E+00	ΣPCBs
A61	NR	M	2007	6.38E+00	ΣPCBs
C22	NR	M	2007	5.56E+00	ΣPCBs
I64	NR	M	2007	2.92E+00	ΣPCBs
I46	NR	M	2007	4.71E-01	ΣPCBs
R31	NR	M	2007	1.39E+01	ΣPCBs
R30	NR	M	2007	4.30E+00	ΣPCBs
R43	NR	M	2007	7.54E+00	ΣPCBs
I46	NR	M	2008	9.97E+00	ΣPCBs
I62	NR	M	2008	5.46E+00	ΣPCBs
I64	NR	M	2008	4.44E+00	ΣPCBs
R28	NR	M	2008	5.94E+00	ΣPCBs
R43	NR	M	2008	8.65E+00	ΣPCBs
R30	NR	M	2008	6.68E+00	ΣPCBs
I42	NR	M	2008	9.52E+00	ΣPCBs
I78	NR	M	2008	5.55E+00	ΣPCBs
I67	NR	M	2008	6.12E+00	ΣPCBs
A39	NR	M	2008	5.01E+00	ΣPCBs
Unknown	NR	M	1993	7.00E-02	ΣPBDEs
Unknown	NR	M	1993	1.52E-01	ΣPBDEs
Unknown	NR	M	1993	7.39E-02	ΣPBDEs
Unknown	NR	M	1993	3.31E-02	ΣPBDEs
Unknown	NR	M	1993	2.49E-02	ΣPBDEs
Unknown	NR	M	1993	6.38E-02	ΣPBDEs
Unknown	NR	M	1993	2.09E-01	ΣPBDEs
Unknown	NR	M	1993	6.84E-02	ΣPBDEs
Unknown	NR	M	1993	8.40E-02	ΣPBDEs
Unknown	NR	M	1993	5.05E-01	ΣPBDEs
Unknown	NR	M	1993	1.79E-02	ΣPBDEs

Animal ID	Ecotype	Sex	Year	Contaminant/TRV	Contaminant
Unknown	NR	M	1994	3.43E-01	ΣPBDEs
Unknown	NR	M	1994	1.16E-01	ΣPBDEs
A55	NR	M	2003	8.27E-02	ΣPBDEs
A60	NR	M	2003	9.83E-01	ΣPBDEs
A33	NR	M	2003	2.02E-01	ΣPBDEs
I67	NR	M	2003	5.11E-01	ΣPBDEs
I42	NR	M	2003	3.23E-01	ΣPBDEs
A74	NR	M	2003	5.69E-01	ΣPBDEs
R28	NR	M	2003	1.58E-01	ΣPBDEs
A71	NR	M	2004	3.28E-01	ΣPBDEs
A71	NR	M	2007	1.14E+00	ΣPBDEs
A61	NR	M	2007	9.35E-01	ΣPBDEs
C22	NR	M	2007	1.01E+00	ΣPBDEs
I64	NR	M	2007	5.10E-01	ΣPBDEs
I46	NR	M	2007	2.18E-01	ΣPBDEs
R31	NR	M	2007	5.17E-01	ΣPBDEs
R30	NR	M	2007	3.08E-01	ΣPBDEs
J6	SR	M	1993	3.87E+00	ΣPCBs
J3	SR	M	1993	1.06E+02	ΣPCBs
J1	SR	M	1996	1.26E+02	ΣPCBs
J18	SR	M	1996	4.12E+01	ΣPCBs
J18	SR	M	2000	1.62E+02	ΣPCBs
L78	SR	M	2004	5.56E+00	ΣPCBs
L74	SR	M	2004	1.45E+01	ΣPCBs
L71	SR	M	2004	1.16E+01	ΣPCBs
L71	SR	M	2004	2.35E+01	ΣPCBs
L74	SR	M	2004	2.94E+01	ΣPCBs
L78	SR	M	2004	1.44E+01	ΣPCBs
J39	SR	M	2006	2.22E+01	ΣPCBs
J1	SR	M	2006	1.17E+02	ΣPCBs

Animal ID	Ecotype	Sex	Year	Contaminant/TRV	Contaminant
J27	SR	M	2006	4.83E+01	ΣPCBs
L57	SR	M	2006	3.65E+01	ΣPCBs
L85	SR	M	2006	3.26E+01	ΣPCBs
J38	SR	M	2007	2.67E+01	ΣPCBs
K21	SR	M	2007	2.48E+01	ΣPCBs
K34	SR	M	2007	2.54E+01	ΣPCBs
L73	SR	M	2007	2.09E+01	ΣPCBs
L87	SR	M	2007	1.57E+01	ΣPCBs
L 116	SR	M	2016	3.10E+01	ΣPCBs
K 25	SR	M	2016	6.71E+00	ΣPCBs
J 49	SR	M	2016	1.81E+01	ΣPCBs
Unknown	SR	M	1993	2.07E-01	ΣPBDEs
Unknown	SR	M	1993	8.33E-01	ΣPBDEs
Unknown	SR	M	1995	1.62E-01	ΣPBDEs
Unknown	SR	M	1995	7.31E-01	ΣPBDEs
Unknown	SR	M	1995	1.21E+00	ΣPBDEs
L78	SR	M	2004	7.10E-01	ΣPBDEs
L74	SR	M	2004	9.38E-01	ΣPBDEs
L71	SR	M	2004	8.57E-01	ΣPBDEs
L71	SR	M	2004	1.73E+00	ΣPBDEs
L74	SR	M	2004	2.07E+00	ΣPBDEs
L78	SR	M	2004	1.73E+00	ΣPBDEs
J39	SR	M	2006	1.00E+01	ΣPBDEs
J1	SR	M	2006	4.53E+00	ΣPBDEs
J27	SR	M	2006	4.20E+00	ΣPBDEs
L57	SR	M	2006	2.20E+00	ΣPBDEs
L85	SR	M	2006	1.67E+00	ΣPBDEs
J37	SR	M	2016	3.29E-01	ΣPBDEs
K25	SR	M	2016	1.05E+00	ΣPBDEs
L72	SR	M	2016	4.96E-01	ΣPBDEs

Animal ID	Ecotype	Sex	Year	Contaminant/TRV	Contaminant
T138	Bigg's	M	1993	2.06E+00	ΣPCBs
T44	Bigg's	M	1996	1.76E+00	ΣPCBs
T54	Bigg's	M	1996	2.15E+00	ΣPCBs
T142	Bigg's	M	1996	2.07E+00	ΣPCBs
T12A	Bigg's	M	1996	3.39E-01	ΣPCBs
T29	Bigg's	M	1996	2.22E+00	ΣPCBs
T162	Bigg's	M	2002	1.63E+00	ΣPCBs
T101A	Bigg's	M	2003	1.40E+00	ΣPCBs
T74	Bigg's	M	2003	1.65E+00	ΣPCBs
T055A	Bigg's	M	2007	1.46E+00	ΣPCBs
TO69C	Bigg's	M	2007	1.79E+00	ΣPCBs
T150	Bigg's	M	2008	1.76E+00	ΣPCBs
T060D	Bigg's	M	2008	2.14E+00	ΣPCBs
T049A	Bigg's	M	2008	1.55E+00	ΣPCBs
T087	Bigg's	M	2008	2.21E+00	ΣPCBs
T090	Bigg's	F	2008	1.66E+00	ΣPCBs
Unknown	Bigg's	M	1993	1.76E-01	ΣPBDEs
Unknown	Bigg's	M	1994	-7.24E-01	ΣPBDEs
Unknown	Bigg's	M	1994	-4.84E-01	ΣPBDEs
Unknown	Bigg's	M	1996	-2.15E-01	ΣPBDEs
Unknown	Bigg's	M	1996	-4.20E-01	ΣPBDEs
Unknown	Bigg's	M	1996	2.15E-02	ΣPBDEs
T101A	Bigg's	M	2003	2.85E-02	ΣPBDEs
T74	Bigg's	M	2003	4.96E-02	ΣPBDEs
T055A	Bigg's	M	2007	-2.95E-02	ΣPBDEs
TO69C	Bigg's	M	2007	5.63E-01	ΣPBDEs

Table H.4 **Ratios of the concentrations of polychlorinated biphenyls (PCBs) found in individual male killer whales to the estimated tissue residue guideline value for PCBs (0.05 mg·kg^{-1} wet weight). Ratios of the concentrations of polybrominated diphenyl ethers (PBDEs) found in individual male killer whales to the corresponding Canadian guidelines for wildlife diet are also given. The source and year of collection for each sample are given.**

Source	Year	Contaminant	Contaminant Concentration/Guideline
Cullon et al. 2009	2000	PCB	1.96E-01
Cullon et al. 2009	2000	PCB	9.50E-02
Cullon et al. 2009	2000	PCB	1.35E-01
Cullon et al. 2009	2000	PCB	3.45E-01
Cullon et al. 2009	2000	PCB	2.66E-01
Cullon et al. 2009	2000	PCB	1.45E-01
Cullon et al. 2009	2000	PCB	6.85E-01
Cullon et al. 2009	2000	PCB	9.18E-01
Cullon et al. 2009	2000	PCB	7.60E-01
Cullon et al. 2009	2000	PCB	1.62E+00
Cullon et al. 2009	2000	PCB	4.36E-01
Cullon et al. 2009	2000	PCB	1.07E+00
Present Study	2014	PCB	4.39E-01
Present Study	2014	PCB	3.72E-01
Present Study	2014	PCB	3.02E-01
Present Study	2014	PCB	4.22E-01
Present Study	2014	PCB	2.70E-01
Present Study	2014	PCB	4.14E-01
Present Study	2014	PCB	6.66E-01
Unpublished Axys Data	2002	PBDE	1.87E+00
Present Study	2014	PBDE	6.25E-01
Present Study	2014	PBDE	4.89E-01
Present Study	2014	PBDE	5.47E-01
Present Study	2014	PBDE	4.38E-01
Present Study	2014	PBDE	2.71E-01

Source	Year	Contaminant	Contaminant Concentration/Guideline
Present Study	2014	PBDE	4.82E-01
Present Study	2014	PBDE	6.41E-01

Table H.5 Individual ratio outcomes for concentrations of polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and isomers of hexabromocyclododecane (HBCDDs) found in Chinook salmon samples collected in 2014, divided by their corresponding guidelines.

Source	Sample	Year	Contaminant	Contaminant Concentration/Guideline
Present Study	Chinook Sample 1	2014	PCB	4.39E-01
Present Study	Chinook Sample 2	2014	PCB	3.72E-01
Present Study	Chinook Sample 3	2014	PCB	3.02E-01
Present Study	Chinook Sample 4	2014	PCB	4.22E-01
Present Study	Chinook Sample 5	2014	PCB	2.70E-01
Present Study	Chinook Sample 6	2014	PCB	4.14E-01
Present Study	Chinook Sample 7	2014	PCB	6.66E-01
Present Study	Chinook Sample 1	2014	PBDE	6.25E-01
Present Study	Chinook Sample 2	2014	PBDE	4.89E-01
Present Study	Chinook Sample 3	2014	PBDE	5.47E-01
Present Study	Chinook Sample 4	2014	PBDE	4.38E-01
Present Study	Chinook Sample 5	2014	PBDE	2.71E-01
Present Study	Chinook Sample 6	2014	PBDE	4.82E-01
Present Study	Chinook Sample 7	2014	PBDE	6.41E-01
Present Study	Chinook Sample 1	2014	HBCDD	1.27E-05
Present Study	Chinook Sample 2	2014	HBCDD	1.26E-05
Present Study	Chinook Sample 3	2014	HBCDD	1.04E-05
Present Study	Chinook Sample 4	2014	HBCDD	1.48E-05
Present Study	Chinook Sample 5	2014	HBCDD	1.25E-05
Present Study	Chinook Sample 6	2014	HBCDD	1.39E-05
Present Study	Chinook Sample 7	2014	HBCDD	1.45E-05

Appendix I.

Results for Contaminants Divided by TRVs

The geometric mean concentrations of contaminants in female killer whales divided by toxicity reference values (TRVs) over time are shown in Figures I.1 – I.3. Linear regression analyses were conducted for the ratio of geometric mean concentrations of each contaminant to its related TRV over time. No significant relationships were found between ΣPCBs/TRV and time, or between ΣPBDEs/TRV and time. The values for the geometric means of each contaminant/TRV for each sample year are presented in Table I.1.

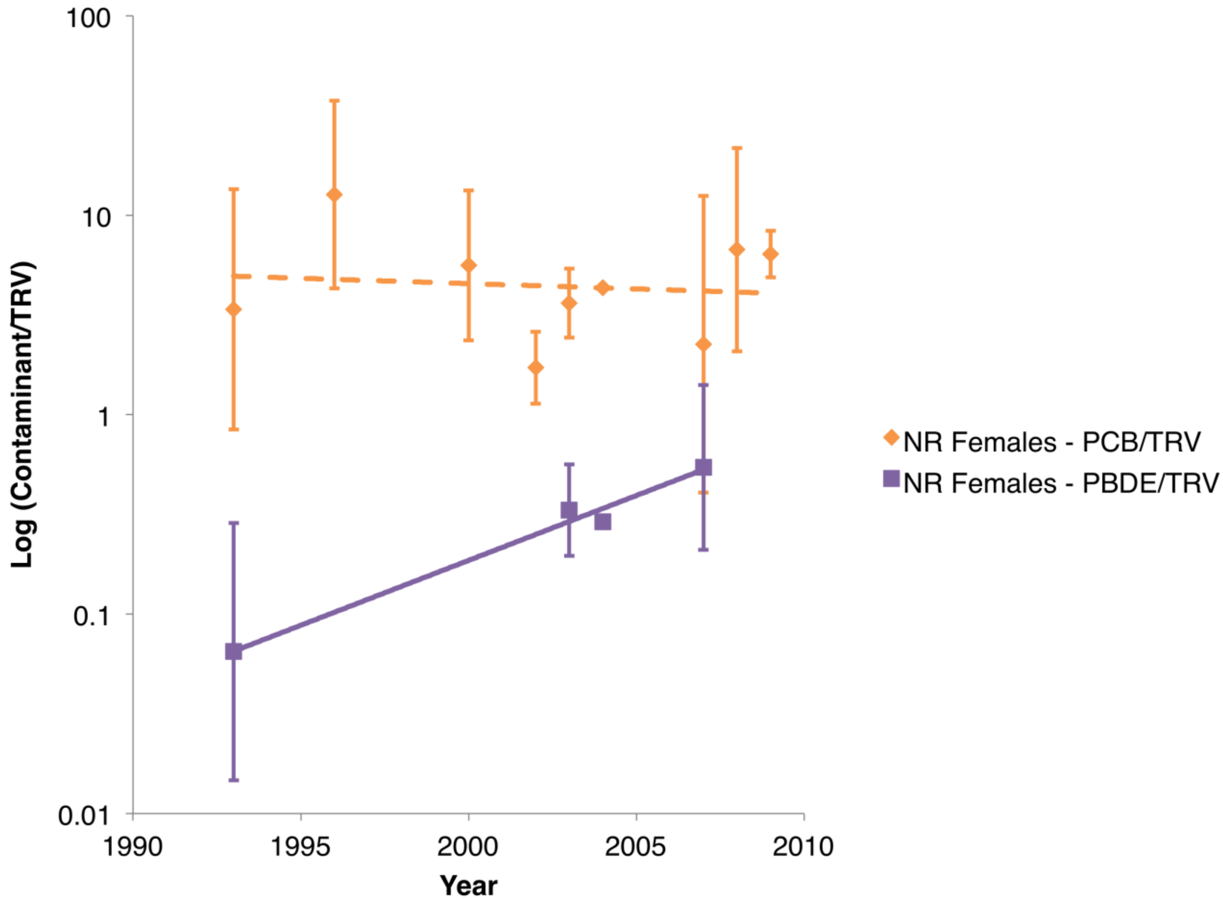

Figure I.1 **The log-transformed geometric means of two contaminants – polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE) – found in female Northern resident (NR) killer whales divided by the corresponding marine mammal toxicity reference value (TRV), plotted against sample year. Concentrations of total PCBs (ΣPCBs) were divided by the average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw), and concentrations of total PBDEs (ΣPBDEs) were divided by the average ΣPBDE TRV of 1.5 mg·kg^{-1} lw. The dashed line represents the linear regression of the geometric means of ΣPCBs/TRV over time, and the solid line represents the linear regression of the geometric means of ΣPBDEs/TRV over time. Error bars represent the standard deviations. If error bars are not shown, it is due to there being insufficient data to calculate the standard deviation for that sample year.**

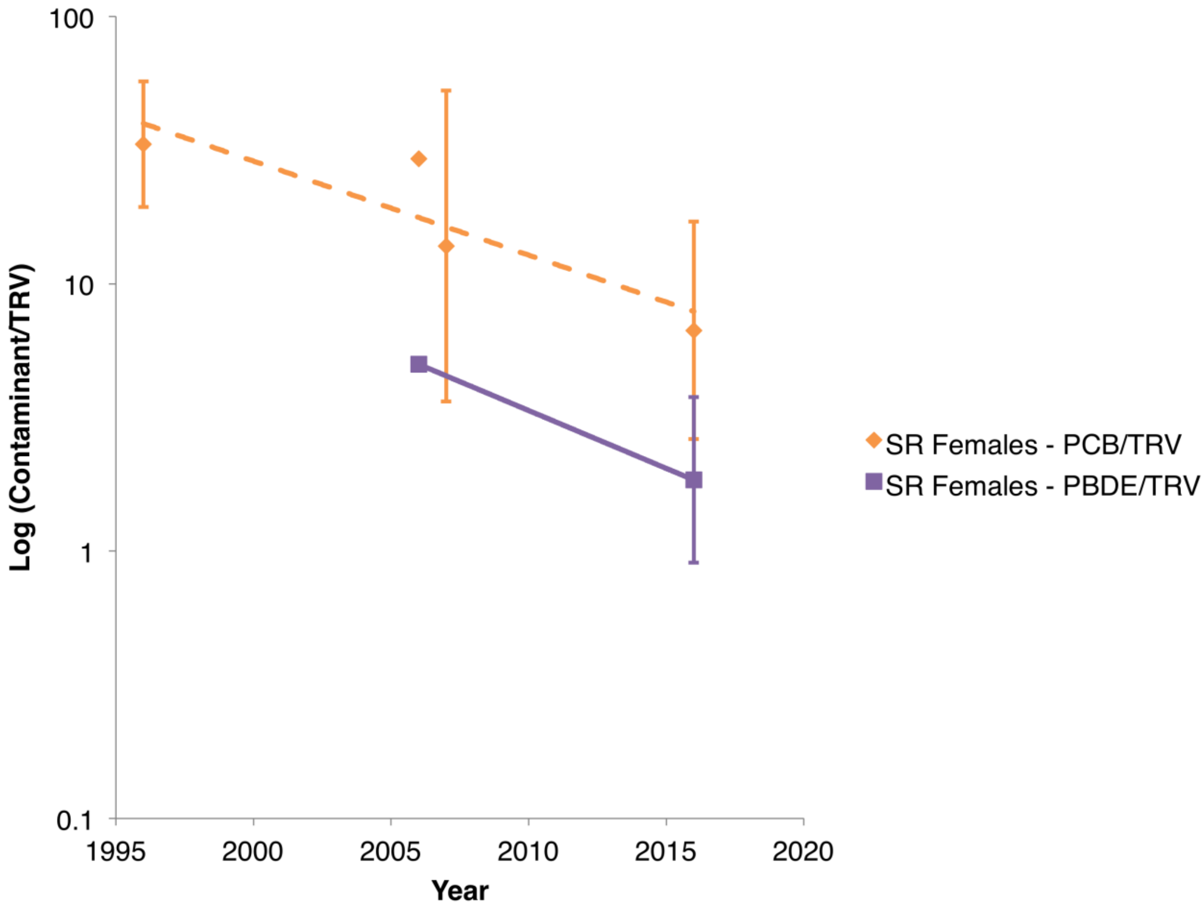

Figure I.2 **The log-transformed geometric means of two contaminants – polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE) – found in female Southern resident (SR) killer whales divided by the corresponding marine mammal toxicity reference value (TRV), plotted against the sample year. Concentrations of total PCBs (ΣPCBs) were divided by the average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw), and concentrations of total PBDEs (ΣPBDEs) were divided by the average ΣPBDE TRV of 1.5 mg·kg^{-1} lw. The dashed line represents the linear regression of the geometric means of ΣPCBs/TRV over time, and the solid line represents the linear regression of the geometric means of ΣPBDEs/TRV over time. Error bars represent the standard deviations. If error bars are not shown, it is due to there being insufficient data to calculate the standard deviation for that sample year.**

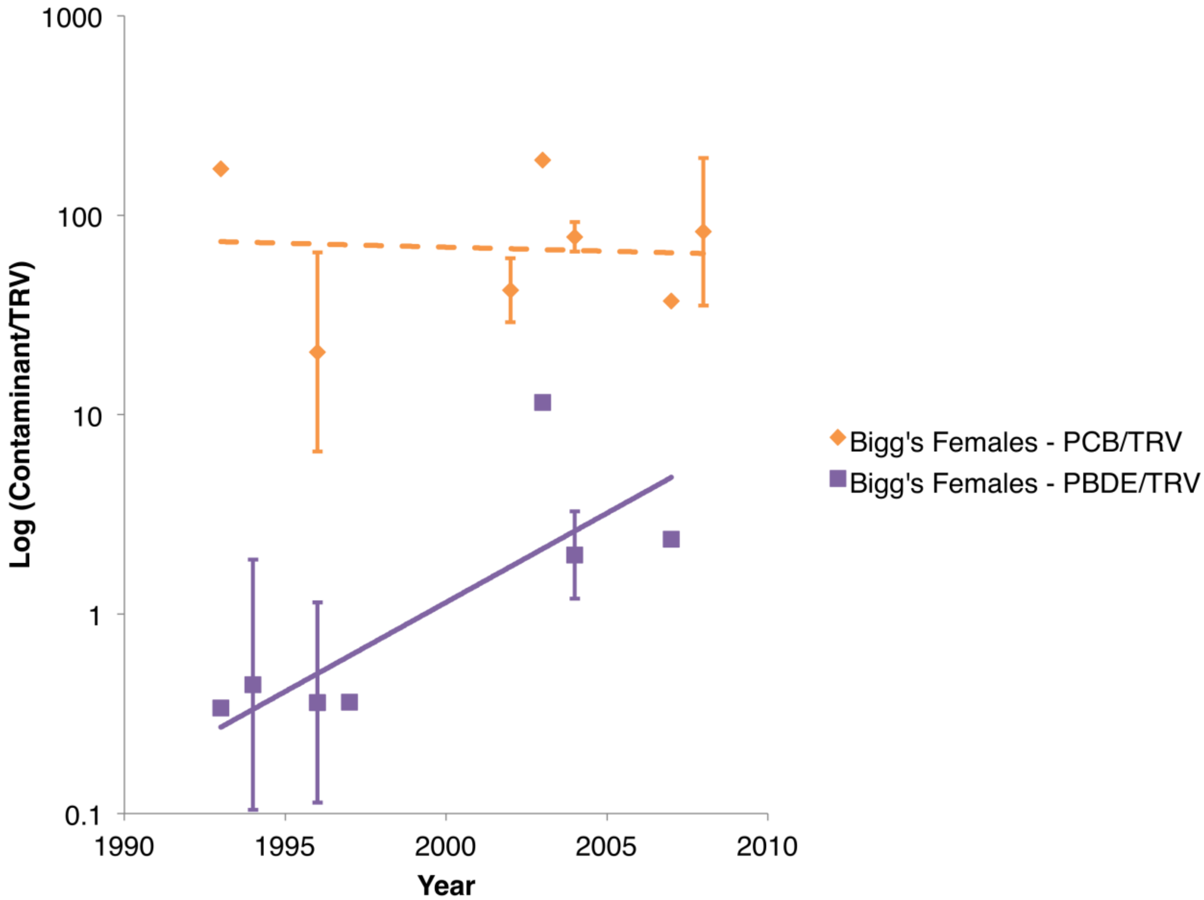

Figure I.3 **The log-transformed geometric means of two contaminants – polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE) – found in female Bigg's killer whales divided by the corresponding marine mammal toxicity reference value (TRV), plotted against the sample year. Concentrations of total PCBs (ΣPCBs) were divided by the average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw), and concentrations of total PBDEs (ΣPBDEs) were divided by the average ΣPBDE TRV of 1.5 mg·kg^{-1} lw. The dashed line represents the linear regression of the geometric means of ΣPCBs/TRV over time, and the solid line represents the linear regression of the geometric means of ΣPBDEs/TRV over time. Error bars represent the standard deviations. If error bars are not shown, it is due to there being insufficient data to calculate the standard deviation for that sample year.**

Table I.1 **Geometric means of the ratios of concentrations of total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs) in individual female killer whales divided by the corresponding toxicity reference value (TRV) for each contaminant. Ecotype, sex, year of collection and standard deviations are given for each sample. The average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw) and ΣPBDE TRV of 1.5 mg·kg^{-1} lw were used for ratio calculations. NR = northern resident, SR = southern resident.**

Ecotype	Sex	Year	Contaminant	Geometric Mean of C_{kw}/TRV	Upper SD	Lower SD
SR	F	1996	PCB	3.32E+01	5.71E+01	1.93E+01
SR	F	2006	PCB	2.93E+01	-	-
SR	F	2007	PCB	1.38E+01	5.28E+01	3.63E+00
SR	F	2016	PCB	6.70E+00	1.71E+01	2.63E+00
NR	F	1993	PCB	3.37E+00	1.35E+01	8.41E-01
NR	F	1996	PCB	1.27E+01	3.76E+01	4.30E+00
NR	F	2000	PCB	5.60E+00	1.33E+01	2.36E+00
NR	F	2002	PCB	1.72E+00	2.61E+00	1.13E+00
NR	F	2003	PCB	3.62E+00	5.39E+00	2.43E+00
NR	F	2004	PCB	4.33E+00	-	-
NR	F	2007	PCB	2.26E+00	1.25E+01	4.06E-01
NR	F	2008	PCB	6.72E+00	2.18E+01	2.08E+00
NR	F	2009	PCB	6.39E+00	8.37E+00	4.88E+00
Bigg's	F	1993	PCB	1.71E+02	-	-
Bigg's	F	1996	PCB	2.06E+01	6.50E+01	6.53E+00
Bigg's	F	2002	PCB	4.21E+01	6.09E+01	2.91E+01
Bigg's	F	2003	PCB	1.89E+02	-	-
Bigg's	F	2004	PCB	7.79E+01	9.24E+01	6.57E+01
Bigg's	F	2007	PCB	3.72E+01	-	-
Bigg's	F	2008	PCB	8.27E+01	1.94E+02	3.52E+01
NR	F	1993	PBDE	6.48E-02	2.87E-01	1.47E-02
NR	F	2003	PBDE	3.32E-01	5.64E-01	1.96E-01
NR	F	2004	PBDE	2.91E-01	-	-

Ecotype	Sex	Year	Contaminant	Geometric Mean of C_{kw}/TRV	Upper SD	Lower SD
NR	F	2007	PBDE	5.44E-01	1.41E+00	2.10E-01
SR	F	2006	PBDE	5.00E+00	#DIV/0!	#DIV/0!
SR	F	2016	PBDE	1.85E+00	3.76E+00	9.05E-01
Bigg's	F	1993	PBDE	3.38E-01	-	-
Bigg's	F	1994	PBDE	4.42E-01	1.87E+00	1.04E-01
Bigg's	F	1996	PBDE	3.60E-01	1.14E+00	1.13E-01
Bigg's	F	1996	PBDE	3.60E-01	1.14E+00	1.13E-01
Bigg's	F	1997	PBDE	3.61E-01	-	-
Bigg's	F	2003	PBDE	1.15E+01	-	-
Bigg's	F	2004	PBDE	1.97E+00	3.27E+00	1.19E+00
Bigg's	F	2007	PBDE	2.38E+00	-	-

- = not applicable due to insufficient data

Results of linear regression analyses investigating the relationship between the geometric mean concentrations of the ratios of contaminants/TRVs for female killer whales and time are presented in Table I.2.

Table I.2 **Results of linear regressions investigating the relationship between the geometric mean of the ratio of contaminants in female killer whales to toxicity reference values (ΣPCBs/TRV or ΣPBDEs/TRV) and time. Analyses were conducted for each ecotype of killer whale – southern resident (SR), northern resident (NR) and Bigg's. The contaminants analyzed included total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs). The linear equation, r^2 value and p-value are given for each combination of ecotype and contaminant that was analyzed. No linear regression analysis was conducted for SR female killer whales due to insufficient data.**

Ecotype	Sex	Contaminant	Data Analyzed	Linear Regression	r^2	p-value
SR	F	PCBs	Geometric Mean	Geometric Mean (ΣPCBs/TRV) = 2.74E+03 - 1.36*Year	0.778	0.118
NR	F	PCBs	Geometric Mean	Geometric Mean (ΣPCBs/TRV) = 2.61E+02 - 0.128*Year	0.0436	0.59
Bigg's	F	PCBs	Geometric Mean	Geometric Mean (ΣPCBs/TRV)= 6.06E+03- 2.98*Year	0.0617	0.591
SR	F	PBDEs	Geometric Mean	-	-	-
NR	F	PBDEs	Geometric Mean	Geometric Mean (ΣPBDEs/TRV) = -60.4 + 0.0303*Year	0.879	0.0622
Bigg's	F	PBDEs	Geometric Mean	Geometric Mean (ΣPBDEs/TRV) = -7.67E+02 + 0.385*Year	0.269	0.188

- = no applicable due to insufficient data to calculate the linear regression

The geometric mean concentrations of contaminants in male killer whales divided by toxicity reference values (TRVs) over time are shown in Figures I.4 – I.6. Linear regression analyses were conducted for the ratio of geometric mean concentrations of each contaminant to its related TRV over time. A significant relationship was found between ΣPBDEs/TRV and time in male NR killer whales (SE = 6.94E-03, $p = 0.0322$). The values for the geometric means of each contaminant/TRV for each sample year are presented in Table I.3. Results of linear regression analyses investigating the relationship between the logarithmic ratios of contaminants/TRVs for male killer whales and time are presented in Table I.4.

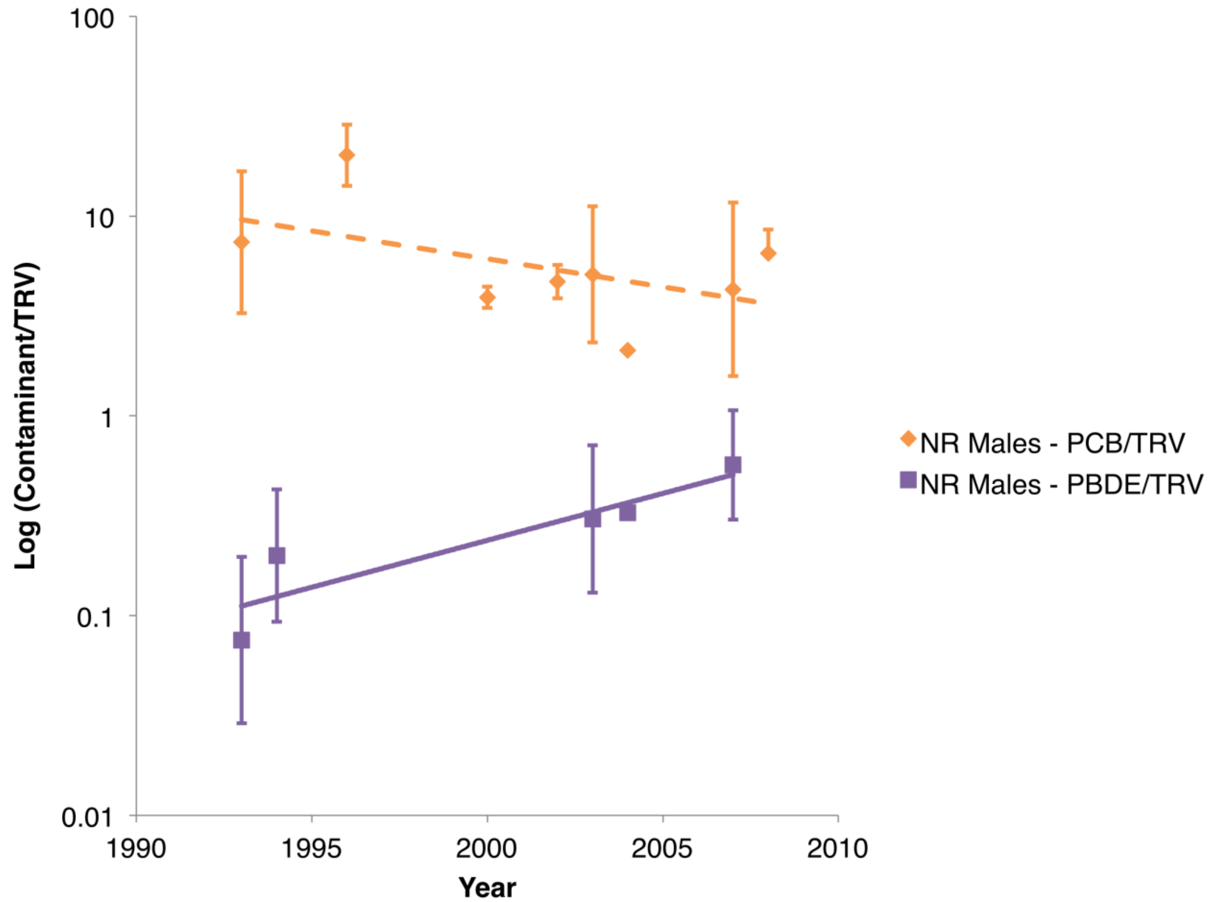

Figure I.4 **The log-transformed geometric means of two contaminants – polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE) – found in male northern resident (NR) killer whales divided by the corresponding marine mammal toxicity reference value (TRV), plotted against sample year. Concentrations of total PCBs (ΣPCBs) were divided by the average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw), and concentrations of total PBDEs (ΣPBDEs) were divided by the average ΣPBDE TRV of 1.5 mg·kg^{-1} lw. The dashed line represents the linear regression of the geometric means of ΣPCBs/TRV over time, and the solid line represents the linear regression of the geometric means of ΣPBDEs/TRV over time. Error bars represent the standard deviations. If error bars are not shown, it is due to there being insufficient data to calculate the standard deviation for that sample year.**

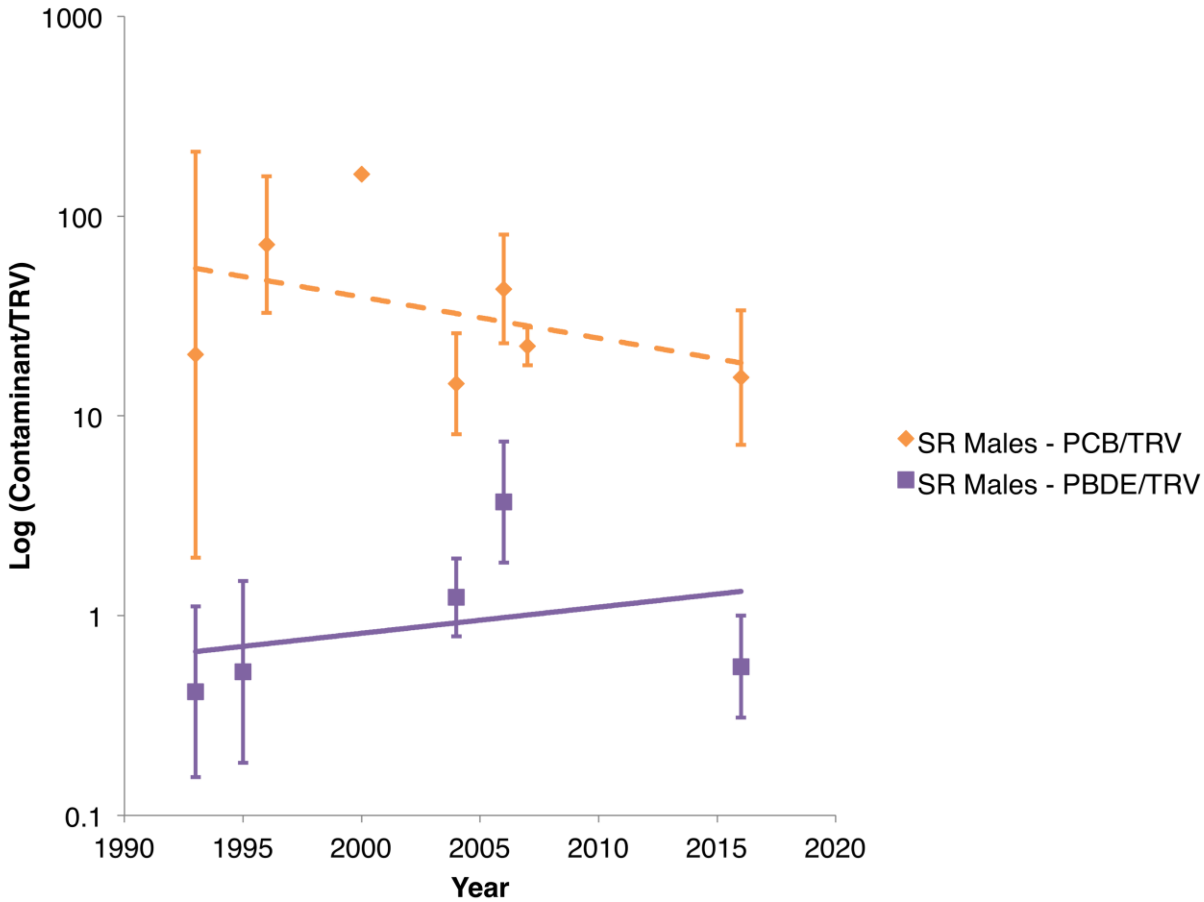

Figure I.5 The log-transformed geometric mean of two contaminants – polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE) – found in male southern resident (SR) killer whales divided by the corresponding marine mammal toxicity reference value (TRV), plotted against sample year. Concentrations of total PCBs (ΣPCBs) were divided by the average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw), and concentrations of total PBDEs (ΣPBDEs) were divided by the average ΣPBDE TRV of 1.5 mg·kg^{-1} lw. The dashed line represents the linear regression of the geometric means of ΣPCBs/TRV over time, and the solid line represents the linear regression of the geometric means of ΣPBDEs/TRV over time. Error bars represent the standard deviations. If error bars are not shown, it is due to there being insufficient data to calculate the standard deviation for that year.

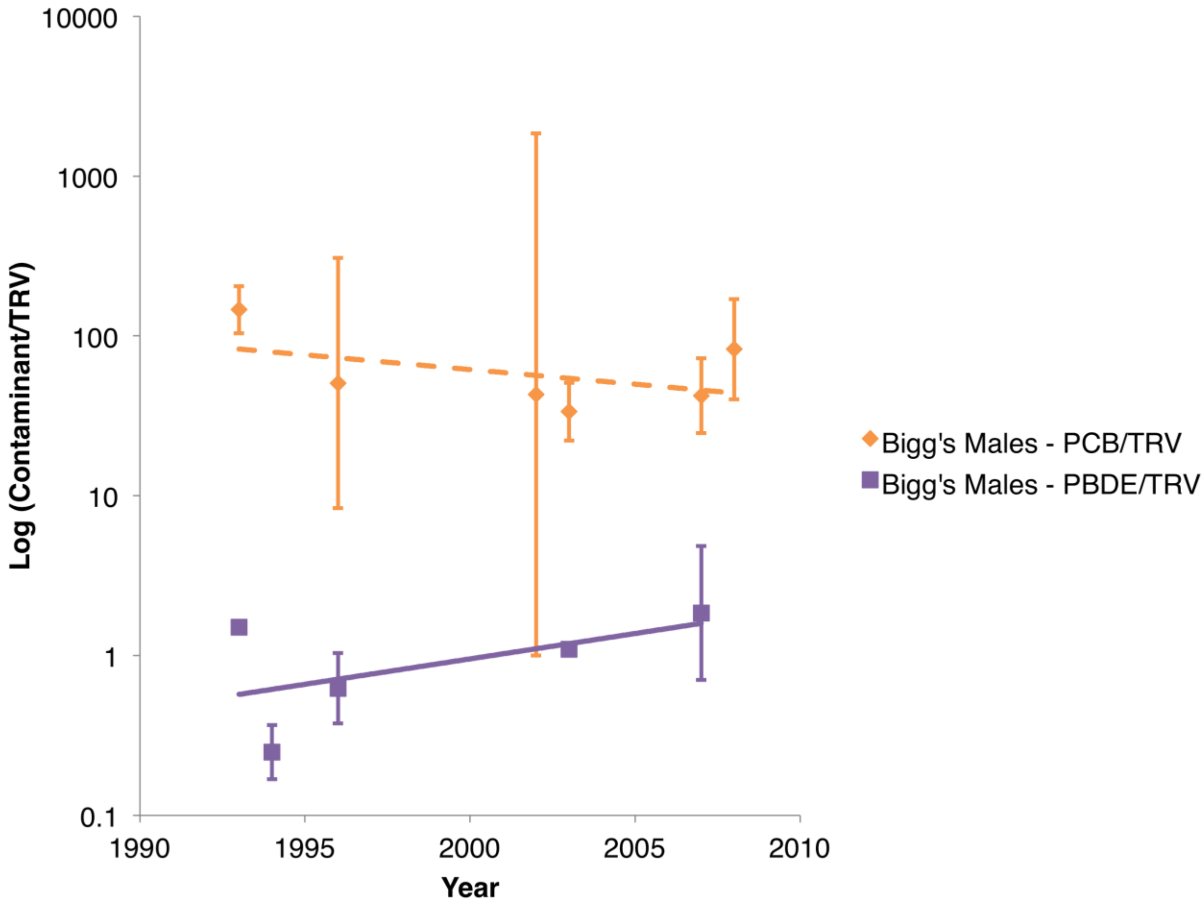

Figure I.6 **The log-transformed geometric means of two contaminants – polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE) – found in male Bigg's killer whales divided by the corresponding marine mammal toxicity reference value (TRV), plotted against sample year. Concentrations of total PCBs (ΣPCBs) were divided by the average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw), and concentrations of total PBDEs (ΣPBDEs) were divided by the average ΣPBDE TRV of 1.5 mg·kg^{-1} lw. The dashed line represents the linear regression of the geometric means of ΣPCBs/TRV over time, and the solid line represents the linear regression of the geometric means of ΣPBDEs/TRV over time. Error bars represent the standard deviations. If error bars are not shown, it is due to there being insufficient data to calculate the standard deviation for that year.**

Table I.3 **Geometric means of the ratios of concentrations of total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs) in individual male killer whales divided by the corresponding toxicity reference value (TRV) for each contaminant. Ecotype, sex, year of collection and standard deviations are given for each sample. The average ΣPCB TRV of 1.5 mg·kg^{-1} lipid weight (lw) and ΣPBDE TRV of 1.5 mg·kg^{-1} lw were used for ratio calculations. NR = northern resident, SR = southern resident.**

Ecotype	Sex	Year	Contaminant	Geometric Mean of C_{kw}/TRV	Upper SD	Lower SD
Bigg's	M	1993	PCB	1.46E+02	2.05E+02	1.04E+02
Bigg's	M	1996	PCB	5.07E+01	3.08E+02	8.34E+00
Bigg's	M	2002	PCB	4.30E+01	1.85E+03	1.00E+00
Bigg's	M	2003	PCB	3.36E+01	5.10E+01	2.21E+01
Bigg's	M	2007	PCB	4.24E+01	7.27E+01	2.47E+01
Bigg's	M	2008	PCB	8.26E+01	1.70E+02	4.01E+01
NR	M	1993	PCB	7.39E+00	1.67E+01	3.26E+00
NR	M	1996	PCB	2.02E+01	2.87E+01	1.42E+01
NR	M	2000	PCB	3.92E+00	4.43E+00	3.46E+00
NR	M	2002	PCB	4.69E+00	5.68E+00	3.87E+00
NR	M	2003	PCB	5.10E+00	1.12E+01	2.33E+00
NR	M	2004	PCB	2.13E+00	-	-
NR	M	2007	PCB	4.30E+00	1.17E+01	1.58E+00
NR	M	2008	PCB	6.50E+00	8.57E+00	4.92E+00
SR	M	1993	PCB	2.02E+01	2.10E+02	1.95E+00
SR	M	1996	PCB	7.19E+01	1.58E+02	3.27E+01
SR	M	2000	PCB	1.62E+02	-	-
SR	M	2004	PCB	1.45E+01	2.59E+01	8.09E+00
SR	M	2006	PCB	4.32E+01	8.06E+01	2.31E+01
SR	M	2007	PCB	2.23E+01	2.77E+01	1.79E+01
SR	M	2016	PCB	1.56E+01	3.38E+01	7.16E+00
NR	M	1993	PBDE	7.54E-02	1.97E-01	2.89E-02
NR	M	1994	PBDE	2.00E-01	4.28E-01	9.32E-02
NR	M	2003	PBDE	3.05E-01	7.13E-01	1.30E-01

NR	M	2004	PBDE	3.28E-01	-	-
NR	M	2007	PBDE	5.68E-01	1.07E+00	3.02E-01
SR	M	1993	PBDE	4.15E-01	1.11E+00	1.55E-01
SR	M	1995	PBDE	5.22E-01	1.49E+00	1.83E-01
SR	M	2004	PBDE	1.23E+00	1.94E+00	7.87E-01
SR	M	2006	PBDE	3.70E+00	7.45E+00	1.84E+00
SR	M	2016	PBDE	5.55E-01	9.99E-01	3.08E-01
Bigg's	M	1993	PBDE	1.50E+00	-	-
Bigg's	M	1994	PBDE	2.49E-01	3.67E-01	1.68E-01
Bigg's	M	1996	PBDE	6.24E-01	1.04E+00	3.75E-01
Bigg's	M	2003	PBDE	1.09E+00	1.13E+00	1.06E+00
Bigg's	M	2007	PBDE	1.85E+00	4.86E+00	7.04E-01

Table I.4 Results of linear regressions investigating the relationship between the geometric mean of the ratio of contaminants in male killer whales to toxicity reference values (ΣPCBs/TRV or ΣPBDEs/TRV) and time. Analyses were conducted for each ecotype of killer whale – southern resident (SR), northern resident (NR) and Bigg's. The contaminants analyzed included total polychlorinated biphenyls (ΣPCBs) and total polybrominated diphenyl ethers (ΣPBDEs). The linear equation, r^2 value and p-value are given for each combination of ecotype and contaminant that was analyzed. No linear regression analysis was conducted for the SR male killer whales due to insufficient data.

Ecotype	Sex	Contaminant	Linear Regression	r^2	p value
Bigg's	M	PCBs	Geometric Mean (PCBs/TRV) = 7.71E+03 - 3.82*Year	0.286	0.274
NR	M	PCBs	Geometric Mean (PCBs/TRV) = 1.14E+03 - 0.566*Year	0.267	0.19
SR	M	PCBs	Geometric Mean (PCBs/TRV) = 4.63E+03 - 2.29*Year	0.107	0.473
Bigg's	M	PBDEs	Geometric Mean (PBDEs/TRV) = -1.22E+02 + 0.0615*Year	0.339	0.303
NR	M	PBDEs	Geometric Mean (PBDEs/TRV) = -52.3 + 0.0263*Year	0.827	0.0322
SR	M	PBDEs	Geometric Mean (PBDEs/TRV) = -71.8 + 0.0365*Year	0.0591	0.694