



Polychlorinated biphenyl and polybrominated diphenyl ether profiles vary with feeding ecology and marine rearing distribution among 10 Chinook salmon (*Oncorhynchus tshawytscha*) stocks in the North Pacific Ocean

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ABSTRACT

Chinook salmon (*Oncorhynchus tshawytscha*) along the west coast of North America have experienced significant declines in abundance and body size over recent decades due to several anthropogenic stressors. Understanding the reasons underlying the relatively high levels of persistent organic pollutants (POPs) in Chinook stocks is an important need, as it informs recovery planning for this foundation species, as well for the Chinook-dependent Resident killer whales (*Orcinus orca*, RKW) of British Columbia (Canada) and Washington State (USA). We evaluated the influence of stock-related differences in feeding ecology, using stable isotopes, and marine rearing ground on the concentrations and patterns of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in Chinook salmon. A principal components analysis (PCA) revealed a clear divergence of PCB and PBDE congener patterns between Chinook with a nearshore rearing distribution ('shelf resident') versus a more offshore distribution. Shelf resident Chinook had 12-fold higher PCB concentrations and 46-fold higher PBDE concentrations relative to offshore stocks. Shelf resident Chinook had PCB and PBDE profiles that were heavier and dominated by more bioaccumulative congeners, respectively. The higher $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in shelf resident Chinook compared to the offshore rearing stocks, and their different marine distributions explain the large divergence in contaminant levels and profiles, with shelf resident stocks being heavily influenced by land-based sources of industrial contamination. Results provide compelling new insight into the drivers of contaminant accumulation in Chinook salmon, raise important questions about the consequences for their health, and explain a major pathway to the heavily POP-contaminated Resident killer whales that consume them.

1. Introduction

Chinook salmon (*Oncorhynchus tshawytscha*) originating from river systems in Canada and the United States (US) on the west coast of North America have experienced precipitous declines in both numbers and size at age over recent decades (Gustafson et al., 2007; Ohlberger et al., 2018; Riddell et al., 2013; Xu et al., 2020), which have been attributed to various anthropogenic stressors, including overfishing, habitat destruction, climate change, and persistent organic pollutants (POPs) (Quinn, 2018). In Canada, the Committee on the Status of Endangered Wildlife in Canada (COSEWIC) determined that 90% (15/16) of the

assessed Fraser River Chinook salmon Conservation Units are at risk for extinction (COSEWIC, 2018). In the US, nine of the 17 Chinook Evolutionary Significant Units (ESU) originating from rivers on the west coast are classified as *Threatened* or *Endangered* under the *Endangered Species Act* (ESA) (Ford, 2022; Waples, 1991; Williams et al., 2016).

Chinook salmon are the largest of the Pacific salmon and are important to Indigenous Peoples, recreational anglers, and commercial fisheries, as well as fish-eating Resident killer whales (RKW, *Orcinus orca*). The *Endangered* Southern Resident killer whales (SRKW) frequent the coastal waters of British Columbia, Canada and Washington state, USA, while the *Threatened* Northern Resident killer whales (NRKW)

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range from Vancouver Island north to southeast Alaska. Both SRKW and NRKW rely on Chinook as their primary prey species with Chinook salmon comprising approximately 80% of the annual SRKW diet and 87% of NRKW diet during the spring and summer (Ford et al., 2010; Hanson et al., 2021). This prey selectivity is further targeted at larger and older Chinook, generally greater than 70 cm or at least 4 years of age (Ford et al., 2010; Ward et al., 2010).

Very high concentrations of POPs, including polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), have been reported in RKWs (Environment and Climate Change Canada, 2020; Ross, 2006; Ross et al., 2000). Chinook salmon have been found to have the highest POP levels among Pacific salmon species, reflecting their longer lifespan and their higher trophic level (Mongillo et al., 2016; Yunker et al., 2011). Such findings have raised important questions about the transport, fate, and effects of food web-amplified POPs. Polychlorinated biphenyls are legacy contaminants that were produced in the US (and other countries) between 1929 and the late 1970s, whereas PBDEs were produced between the late 1970s and 2013. Polychlorinated biphenyls were commercially used as heat-resistant chemicals in electrical transformers, capacitors, and adhesives among other uses (Erickson, 1997), while PBDEs were used widely as flame retardants in electronics, textiles, and household furniture (World Health Organization, 1994). Both PCBs and PBDEs are classes of organic compounds, each with a theoretically possible 209 congeners that reflect the degree of halogenation (chlorine or bromine) on two phenyl rings. Both persist in the environment, bioaccumulate in organisms, biomagnify in food webs, and are toxic (Lallas, 2001; Secretariat of the Stockholm Convention, 2019).

Polychlorinated biphenyls have a lower range of octanol-water partition coefficients ($\log K_{ow}$ 4–8) and take longer to eliminate in fish ($t_{1/2}$ up to > 1000 d), compared to PBDEs, which have a higher $\log K_{ow}$ range (5–10) and are more readily eliminated ($t_{1/2}$ < 500 d) (Di Paolo et al., 2010; Makino, 1998; Niimi and Oliver, 1983; Papa et al., 2009). The 209 theoretically possible congeners of PCBs and PBDEs have provided opportunities to study transport and fate processes for these contaminants. Polychlorinated biphenyl and PBDE profiles measured in the juvenile Chinook salmon, harbor seals (*Phoca vitulina*), and Pacific herring (*Clupea pallasii*) have been associated with their feeding ecology and proximity to pollution hotspots (O'Neill et al., 2020; Ross et al., 2004; West et al., 2008).

Chinook salmon have been shown to accumulate the majority of their POPs during their time at sea (Cullon et al., 2009; O'Neill and West, 2009). However, the extent to which the wide variation in marine spatial distributions of rearing Chinook affect contaminant exposure and accumulation is not clear, with some stocks migrating to rear in the Gulf of Alaska, while others remain closer to their natal rivers and along the continental shelf (i.e., shelf residents) (Fig. 1) (COSEWIC, 2018; Weitkamp, 2010). Resident Chinook that remain in Puget Sound and other urbanized areas of the Salish Sea for their marine rearing were found to have higher POP concentrations than Chinook that migrated to northern rearing grounds (O'Neill and West, 2009; O'Neill et al., 2006).

Carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope ratios have been used extensively to evaluate the role of feeding ecology in fish, and may provide insight into the consequences for contaminant exposure in different Chinook stocks (Fisk et al., 2001). Differences between use of marine (or pelagic) versus coastal (or benthic) carbon food sources can

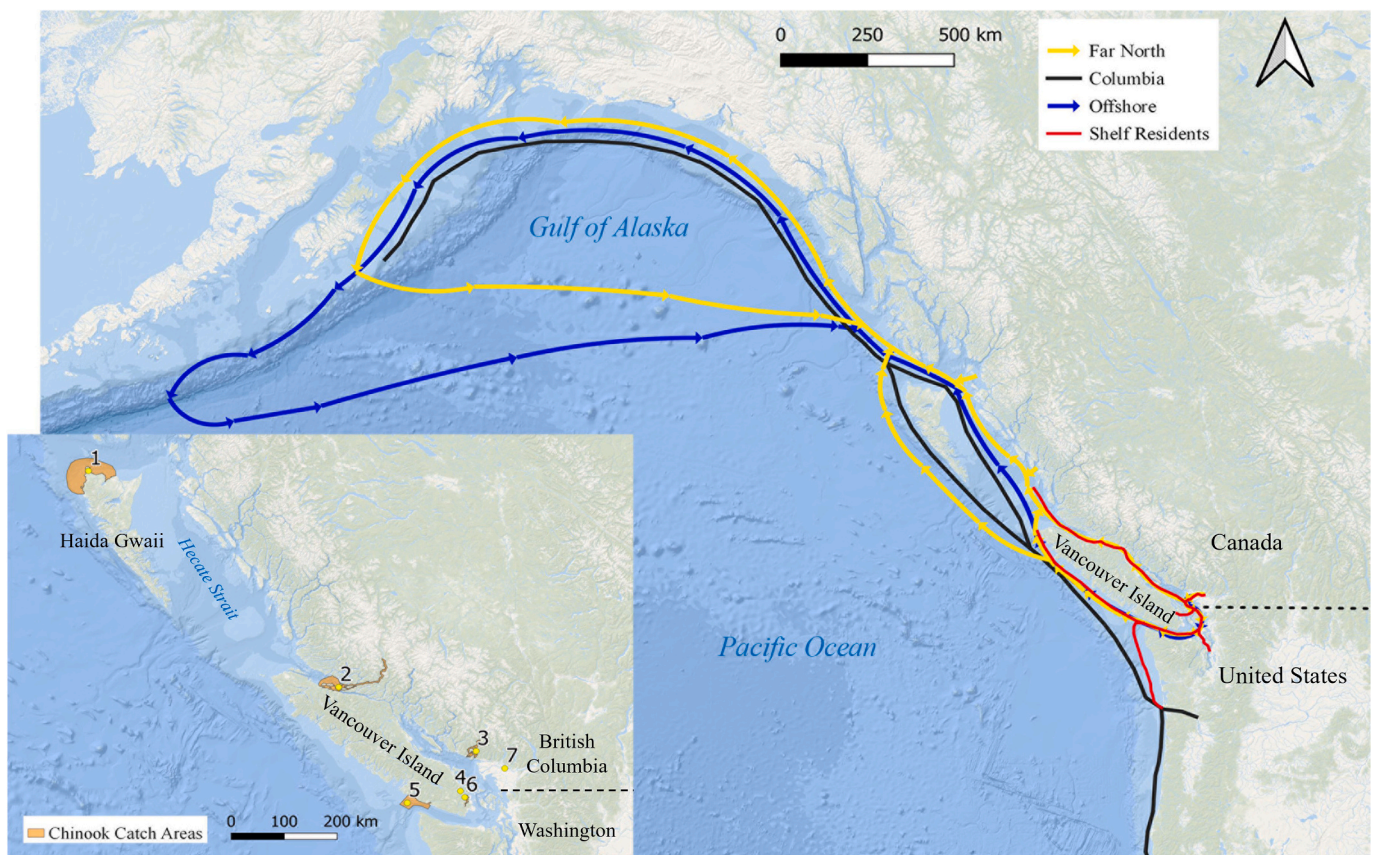


Fig. 1. Estimated marine distribution and rearing grounds of British Columbia (Canada) and US Pacific Northwest immature and maturing Chinook salmon ($n = 83$). Data source: PSC Coded-Wire Tag Program, DFO catch data, and known life history patterns, see Methods section for references. Catch locations by number: 1—NW Coast Haida Gwaii (Langara Island), 2—Blackfish Sound & Knight Inlet, 3—Howe Sound, 4—Cowichan River, 5—Swiftsure Bank, 6—Mill Bay, 7—Lower Fraser Albion Test Fishery. Marine distributions for the various stocks are represented by color-coded lines: Shelf residents (Cowichan, Harrison, Puget Sound), Offshore (Upper & Middle Fraser), Far North (South Thompson, Skeena, North-Central BC, & WCVI), Columbia (Upper & Lower Columbia).

be distinguished with $\delta^{13}\text{C}$, while $\delta^{15}\text{N}$ can provide an indication of trophic level (Das et al., 2004; Zanden and Rasmussen, 2001).

The objective of this study was to characterize the role that feeding ecology and marine rearing strategies play in shaping PCBs and PBDEs in 10 priority stocks of Chinook salmon consumed by RKWs.

2. Methods

2.1. Chinook sample collection and preparation

Adult Chinook salmon were collected in seven areas in the coastal waters off British Columbia, Canada, stretching from the mouth of the Fraser River in the south to the northwest coast of Haida Gwaii (Fig. 1). Collections took place during October 2018 and March–December 2019 in partnership with First Nations and recreational and commercial anglers. Individual Chinook salmon samples (n = 83) from 10 stocks consumed by SRKW and NRKW (Ford et al., 2010; Hanson et al., 2010, 2021) were selected: Cowichan River (East Coast Vancouver Island (ECVI)), West Coast Vancouver Island (WCVI), Puget Sound, Columbia River (Upper and Lower Columbia), Skeena River, North-Central British Columbia, Upper Fraser (Spring 5₂), Middle Fraser (Summer 5₂), South Thompson River (Summer 4₁), and Harrison River (Fall 4₁, Lower Fraser) (Table 1). Marine migration routes and rearing grounds of the selected Chinook stocks (Fig. 1) were estimated using Coded Wire Tagging (CWT), catch data, genotyping, and modeling studies (Beamish et al., 2011; Brown et al., 2019; COSEWIC, 2018; Doutaz et al., 2021; Freshwater et al., 2021; Larson et al., 2013; CTC, 2019; Sharma and Quinn, 2012; Shelton et al., 2019; Tucker et al., 2012; Weitkamp, 2010). Note, Chinook stocks may spend any amount of time along the estimated route and will not necessarily follow the exact path.

Chinook heads were individually bagged and frozen at $-20\text{ }^\circ\text{C}$ and shipped to Fisheries and Oceans Canada (West Vancouver, BC, Canada), where they were stored at $-20\text{ }^\circ\text{C}$. Heads were partially thawed and muscle tissue from the back of the head was subsampled from each individual for stable isotope and contaminant analyses. Subsamples were individually wrapped in acetone-hexane rinsed foil, placed in Whirlpaks®, and stored at $-20\text{ }^\circ\text{C}$.

Biological data including sex, weight, fork length, fin clip for stock identification, and scale for ageing were collected for each individual when possible. Genetic stock and age analysis were performed at Fisheries and Oceans Canada (Nanaimo, BC, Canada). Genetic stock assignment for each Chinook was performed using either cBAYES in conjunction with a baseline of 52,000 individuals from 325 populations derived from microsatellite markers (Beacham et al., 2006) or parentage-based tagging (PBT) and genetic stock identification (GSI) with a baseline of 36,241 individuals from 45 populations derived from 321 single nucleotide polymorphisms (SNPs) (Beacham et al., 2018). Results were reported as Canada Conservation Unit (CU) or United States genetic group and spawning river. As per methods described elsewhere (Beacham et al., 2006, 2018; Ford et al., 2010; Hanson et al., 2010), we combined individual genetic assignments into stocks (see Table S1). To control for age as a confounding factor, adult Chinook aged 4 or 5 years were selected for contaminant and stable isotope analyses. Some individuals could not be aged; therefore we selected Chinook with fork lengths that ranged from 70 to 99 cm, which corresponds with size estimates for 4 or 5 year old adult Chinook (Ford and Ellis, 2006), and is also within the range for size selectivity preferences for RKWs (Ford et al., 2010; Ward et al., 2010).

Despite the Upper Columbia Chinook having a slightly farther north marine rearing strategy (northeast Alaska) than Lower Columbia Chinook, which tend to range farther south to Oregon (Fig. 1, black line) (Shelton et al., 2019; Weitkamp, 2010), their marine rearing areas have considerable overlap (Sharma and Quinn, 2012) and as such were combined into one stock group for biological, dietary, and contaminant analyses. Stable isotope ratios, C:N ratios, $\sum\text{PCBs}$, and $\sum\text{PBDEs}$ did not differ between Upper and Lower Columbia ($\delta^{15}\text{N}$ t-test, $p = 0.4$; $\delta^{13}\text{C}$

Table 1
Morphometric and stable isotope values vary among 10 priority Chinook stocks consumed by Northern and Southern Resident killer whales. ANOVA and Tukey results for fork length showed significant pairwise comparisons for SK > PS ($p = 0.02$) and NB > PS ($p < 0.01$).

Stock	n	Fork Length (cm)	Sex (F:M:U)	Age (Years)	$\delta^{13}\text{C}$ normalized	ANOVA for $\delta^{13}\text{C}$ (Tukey's tests)	$\delta^{15}\text{N}$	ANOVA for $\delta^{15}\text{N}$ (Tukey's tests)
Skeena (SK)	4	91.2 ± 1.0	1:3:0		-18.24 ± 0.58	LF, CO, PS	14.46 ± 0.69	
North-Central BC (NB)	7	89.7 ± 2.9	5:0:2		-17.48 ± 0.21	MF, LF, CO	14.81 ± 0.21	
Upper Fraser (UF)	9	82.0 ± 3.0	5:2:1	4.8 ± 0.1	-18.43 ± 0.15	LF, WV, CO, PS, CL	14.79 ± 0.11	
Mid Fraser (MF)	15	80.6 ± 1.6	7:5:3	4.5 ± 0.1	-18.42 ± 0.13	NB, LF, WV, CO, PS, CL	14.47 ± 0.14	
South Thompson (ST)	12	81.1 ± 1.0	7:5:0	4.1 ± 0.1	-17.90 ± 0.14	LF, CO, PS	14.21 ± 0.15	
Lower Fraser (Harrison) (LF)	12	82.8 ± 2.1	6:6:0	4.1 ± 0.1	-16.28 ± 0.14	SK, NB, UF, MF, ST, WV, CL	15.01 ± 0.16	ST
WCVI (WV)	12	80.1 ± 1.5	6:6:0		-17.25 ± 0.14	UF, MF, LF	14.52 ± 0.13	
Cowichan (ECVI) (CO)	12	81.5 ± 2.0	0:12:0	3.4 ± 0.1	-16.52 ± 0.13	SK, NB, UF, MF, ST, CL	15.51 ± 0.09	SK, MF, ST, WV, PS, CL
Puget Sound (PS)	14	78.1 ± 1.9	9:4:1		-17.02 ± 0.22	SK, UF, MF, ST	14.80 ± 0.12	
Columbia (CL)	15	80.4 ± 1.9	7:7:1		-17.32 ± 0.17	UF, MF, CO, LF	14.45 ± 0.10	

Sex is shown as a ratio of female (F) to male (M) to unknown (U). n represents the number of individuals sampled in each stock. Other values are shown as mean ± standard error. Stock order is based on approximate river location North to South along the west coast of North America.

t -test, $p = 0.4$; C:N t -test, $p = 0.5$; \sum PCBs Mann-Whitney U test, $p = 0.2$; \sum PBDEs Mann-Whitney U test, $p = 0.3$) thus were not separated for analyses. The lack of significant differences could be due to sample size (Table S1), which merits further investigation with greater sample size of both Upper and Lower Columbia Chinook.

2.2. Chemical analyses

Muscle tissue samples ($n = 83$) were analyzed for percent lipid, 159 PCB congeners, and 40 PBDE congeners at SGS AXYS Analytical Services Ltd., Sidney, British Columbia, Canada according to laboratory protocols and criteria described elsewhere (Christensen et al., 2005). Polychlorinated biphenyls and PBDEs were analyzed using high-resolution gas chromatography coupled with high-resolution mass spectrometry (HRGC-HRMS) detection according to the US Environmental Protection Agency (EPA) methods 1668A (1997) and 1614 (2007), respectively. For each batch of up to 15 samples, a method blank and standards were analyzed. Polychlorinated biphenyl and PBDE congener concentrations were blank-corrected and lipid-weight adjusted. Congener measurements below the detection limit were replaced with a random number generated between zero and the detection limit. Total PCB and PBDE concentrations are reported as \sum PCB or \sum PBDE in nanogram/gram lipid weight (ng/g lw).

2.3. Stable isotope analysis

Muscle tissue was oven dried, ground into a powder, weighed (~500 μ g) in tin capsules and analyzed for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ by a Delta V Thermo Scientific Continuous Flow Mass Spectrometer (Thermo Scientific, Bremen, Germany) coupled to a 4010 Elemental Combustion System (Costech Instruments, Valencia, CA, USA). Samples were analyzed at the University of Windsor Great Lakes Institute of Environmental Research (GLIER), Ontario, Canada. Carbon and nitrogen stable isotopes were expressed by δ values in parts per thousand (‰) deviation from the international standards Vienna Pee Dee Belemnite ($\delta^{13}\text{C}$) and atmospheric air ($\delta^{15}\text{N}$). The δ values for carbon and nitrogen stable isotopes were calculated as follows:

$$\delta X\text{‰} = \left[\left(\frac{R_{\text{sample}}}{R_{\text{standard}}} \right) - 1 \right] \times 1000 \quad (1)$$

where X is the isotope being measured and R is the ratio of heavy to light isotope ($^{13}\text{C}/^{12}\text{C}$ or $^{15}\text{N}/^{14}\text{N}$). All $\delta^{13}\text{C}$ values were lipid corrected using the Kiljunen et al. (2006); McConnaughey and McRoy (1979) equations:

$$\delta^{13}\text{C Lipid extracted} = \delta^{13}\text{C} + 7.018 * (0.048 + 3.90/(1 + 287/L)) \quad (2)$$

$$L (\text{percent lipid}) = 93 / \left[1 + (0.246 \text{ C/N} - 0.775)^{-1} \right] \quad (3)$$

where C/N = ratio of percent carbon to percent nitrogen in the sample. This lipid correction was performed because Chinook salmon have relatively high lipid content (C/N > 3.4, Kiljunen et al., 2006), which has been shown to artificially deplete $\delta^{13}\text{C}$ (Post et al., 2007). This lipid correction was recently validated in 2021 (Larocque et al., 2021; Kiljunen et al., 2006; McConnaughey and McRoy, 1979).

Precision was evaluated using the standard deviation of replicate analyses of four standards: NIST1577c, internal lab standard (tilapia muscle), USGA 40 and Urea ($n = 42$). Standard deviation for all standards was $\leq 0.24\text{‰}$ for $\delta^{15}\text{N}$ and $\leq 0.22\text{‰}$ for $\delta^{13}\text{C}$. $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of USGA 40 ($n = 50$ for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ each) analyzed throughout runs of samples showed a difference of 0.01‰ for $\delta^{15}\text{N}$ and 0.19‰ for $\delta^{13}\text{C}$ from the certified values. The accuracy of the instrumentation was based on NIST standards 8573, 8547, and 8574 for $\delta^{15}\text{N}$ and 8542, 8573, and 8574 for $\delta^{13}\text{C}$ ($n = 10$ for all). The mean difference from the certified values were 0.02 , 0.22 , and 0.20‰ for $\delta^{15}\text{N}$ and 0.14 , -0.23 , and -0.21‰ for $\delta^{13}\text{C}$, respectively.

2.4. Statistical analysis

All statistical analyses were carried out using R Statistical Software (Ver 4.0.2: R Foundation for Statistical Computing) (R Core Team, 2020) with statistical significance set as $\alpha = 0.05$. Maps were produced using QGIS 3.16.3 Hanover. The Shapiro-Wilk test was used to test the normality and homogeneity of variances for contaminant concentrations and stable isotopes variables. Variables that deviated from the normal distribution even after \log_{10} transformation were analyzed using non-parametric tests. Mann-Whitney U tests were performed to test for differences in \sum PCB and \sum PBDE concentrations between Upper and Lower Columbia Chinook. T-tests were used to compare stable isotope ratios between Upper and Lower Columbia Chinook. T-tests were also used to compare stable isotopes ratios and lipid content between male and female Chinook. Two-way analysis of variance (ANOVA) was used to assess the effect of stock and sex on lipid content. Because Cowichan, North-Central BC, and Skeena Chinook did not have an even distribution of male and females, they were not included in the two-way ANOVA that included sex as a factor. One-way ANOVA was used to compare the means of stable isotopes, fork length, and percent lipid among Chinook stocks. Significant ANOVAs were followed with Tukey's post hoc tests to determine specific differences among Chinook stocks. A Kruskal-Wallis test followed by a pairwise Wilcoxon rank sum test with Benjamini-Hochberg correction was used to compare \sum PCB or \sum PBDE concentrations among Chinook stocks.

Principal component analysis (PCA) was performed to investigate differences in PCB and PBDE congener patterns in Chinook stocks. Congeners not detected in at least 70% of samples were removed. Data was standardized to the concentration total before the PCA to eliminate concentration bias between samples. The centered \log_{10} ratio transformation (each congener normalized by the geomean followed by a \log_{10} transformation) was then applied to produce a data set unaffected by negative bias or closure (Aitchison, 1986). The data were then auto scaled (scaled to variable mean and standard deviation) before input into the PCA.

Linear regressions were used to assess the relationship between PCB and PBDE PCA projections and biological variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, percent lipid, stock). An Akaike Information Criteria (AICc) calculations correcting for small sample sizes (Burnham, 2002) in conjunction with a backward stepwise regression was used to assess which variable or combination of variables best explained the variation observed in the PCB and PBDE Chinook patterns.

The comprehensive dataset of detected PCBs and PBDEs (127 congeners) for the adult Chinook afforded the opportunity to use cluster analysis to further explore the congener patterns for these two important contaminant classes. An exploratory cluster analysis was conducted using the PCB and PBDE data for all Chinook to further characterize the patterns and groupings across the various stocks. K-means and hierarchical cluster analysis using Ward's minimum variance method were used on combined PCB and PBDE congener data to assess groupings of Chinook stocks. While both k-means (non-hierarchical) and hierarchical clustering divide data points into groups based on their similarity, k-means uses a pre-defined number of groups denoted as "K" to create clusters, whereas hierarchical clustering assembles the dataset into dendrograms to create a tiered structure of clusters.

3. Results and discussion

3.1. Biological and dietary variations among Chinook stocks

Mean lipid content in Chinook muscle varied among the different stocks (ANOVA; $F_{9,73} = 20.9$, $p < 0.001$). Cowichan Chinook exhibited lower mean lipid content ($2.49 \pm 0.26\%$) compared to all other stocks ($p < 0.05$), except for Harrison ($5.86 \pm 0.79\%$; $p = 0.3$) which had the second lowest lipid content followed by WCVI Chinook (Tables S8 and S3). Mean lipid content in Harrison Chinook was lower than the Upper

and Mid Fraser (16.6 ± 0.8 and $15.6 \pm 0.9\%$, respectively) and South Thompson ($13.9 \pm 0.9\%$) stocks ($p < 0.001$, Table S3). These three latter stocks had the highest mean lipid content across all stocks (Table S8).

These findings are consistent with the different energy needs of Chinook stocks undertaking their respective freshwater migration to natal spawning grounds. For example, Upper and Mid Fraser Chinook, along with South Thompson stocks, have high lipid content to fuel long (705–994 km) and demanding migrations (606–713 m in elevation) once they enter the Fraser River (COSEWIC, 2018; Lerner and Hunt, 2023). Cowichan and Harrison Chinook have lower lipid content, reflecting their shorter freshwater migrations. Our results are also consistent with previous studies that showed Harrison Chinook have the lowest percent lipid content relative to South Thompson and Upper and Mid Fraser stocks (Cullon et al., 2009; Lerner and Hunt, 2023; O'Neill et al., 2014; Veldhoen et al., 2010). Percent lipid values were negatively correlated with $\delta^{13}\text{C}$ ($R^2 = -0.393$; $p < 0.001$) and $\delta^{15}\text{N}$ ($R^2 = -0.206$; $p < 0.001$). This relationship between percent lipid and $\delta^{13}\text{C}$ supports our results; shelf resident Chinook stocks (i.e., Harrison and Cowichan) residing and foraging in coastal waters have lower lipid reserves that reflect the shorter distances back to their spawning grounds, whereas offshore rearing stocks with longer return migrations have higher lipid reserves.

The lipid content variability among stocks, particularly for Harrison and Cowichan caught in close proximity to their natal rivers, may be influenced to some extent by sexual maturation, a period when Chinook lipid reserves are mobilized from muscle to gonads (Hendry and Berg, 1999). Chinook arrive at freshwater entry at varying levels of maturity due to their different spawning times (Hearsey and Kinziger, 2015). So it is possible that the fall run of Cowichan and Harrison Chinook in the present study may have started to sexually mature. However, a recent study examining Harrison Chinook caught from the same location as in the lower Fraser River did not find significant mobilization of muscle lipids to gonads (Lerner and Hunt, 2023).

Mean lipid content was not significantly different in females ($n = 53$; $10.1 \pm 4.6\%$) and males ($n = 51$; $8.35 \pm 5.0\%$) ($p = 0.07$). Further, using a two-way ANOVA we tested for the effect of stock and sex on lipid content, and while a significant effect based on stock was found ($F_{6,66} = 10.4$, $p < 0.001$), there was no effect from sex or the interaction between stock and sex ($p > 0.05$).

While age data was not available for our Skeena and North-Central Chinook, their size ranged from 89 to 94 cm and 79 to 99 cm (Table S1), respectively. Skeena (SK) and North-Central BC (NB)

Chinook had a greater mean fork length (cm) than Puget Sound Chinook (Table 1; ANOVA; $F_{9,98} = 2.93$, $p = 0.004$; NB: $p < 0.01$, SK: $p = 0.02$). A previous comparison between Skeena and Puget Sound Chinook also showed Skeena Chinook had greater mean fork length than Puget Sound Chinook (O'Neill et al., 2014). No other differences in fork length ($p > 0.05$) were found among the Chinook stocks, which differs from recent findings that showed fork length differences among Fraser River Chinook (Lerner and Hunt, 2023). However, the latter study did not indicate if age was controlled for when performing their length comparisons. No significant differences were found between males and females for fork length ($p = 0.3$).

Mean $\delta^{13}\text{C}$ varied among the 10 Chinook stocks (ANOVA; $\delta^{13}\text{C } F_{9,102} = 18.1$, $p < 0.001$) (Table 1; Fig. 2). Mean $\delta^{13}\text{C}$ in shelf resident Cowichan Chinook were higher compared to Upper and Mid Fraser, Columbia, Skeena, South Thompson, and North-Central BC Chinook ($p < 0.05$, Fig. 2). Mean $\delta^{13}\text{C}$ in shelf resident Harrison Chinook were higher compared to those in Upper and Mid Fraser, Columbia, South Thompson, Skeena, West Coast Vancouver Island, and North-Central BC Chinook ($p < 0.05$). Puget Sound Chinook, which are also a shelf rearing Chinook stock had higher mean $\delta^{13}\text{C}$ compared to those in Upper and Mid Fraser, South Thompson, and Skeena Chinook ($p < 0.05$). The higher $\delta^{13}\text{C}$ in the three shelf resident Chinook stocks (Cowichan, Harrison, and Puget Sound) reflect their use of the coastal shelf habitats (Beamish et al., 2011; Doutaz et al., 2021; Shelton et al., 2019), relative to the other stocks, such as the Upper and Mid Fraser Chinook, which have lower $\delta^{13}\text{C}$ reflecting their use of offshore food webs (Doutaz et al., 2021) (Figs. 1 and 2). Cowichan Chinook remain relatively close to their natal river rearing along the east coast of Vancouver Island and around the Gulf Islands in the Strait of Georgia (Beamish et al., 2011; Shelton et al., 2019). Harrison Chinook rear in the Salish Sea or the coastal waters of Washington and Oregon within 1000 km from the mouth of the Fraser River (Doutaz et al., 2021). Similarly, Puget Sound Chinook tend to utilize more coastal shelf habitats migrating on either side of Vancouver Island but not much farther than north Vancouver Island (Freshwater et al., 2021; Shelton et al., 2019; Weitkamp, 2010), with a small percentage of stocks (5–10% of the 2018 and 2019 southeast Alaska Chinook troll fishery catch) migrating as far as southeast Alaska (Shedd et al., 2021, 2022).

Mean $\delta^{13}\text{C}$ of Columbia River, WCVI and North-Central BC were higher than those of Upper and Mid Fraser Chinook ($p < 0.05$), but the relationship between North-Central BC and Upper Fraser was marginally significant ($p = 0.05$). These relationships reflect the north and far north

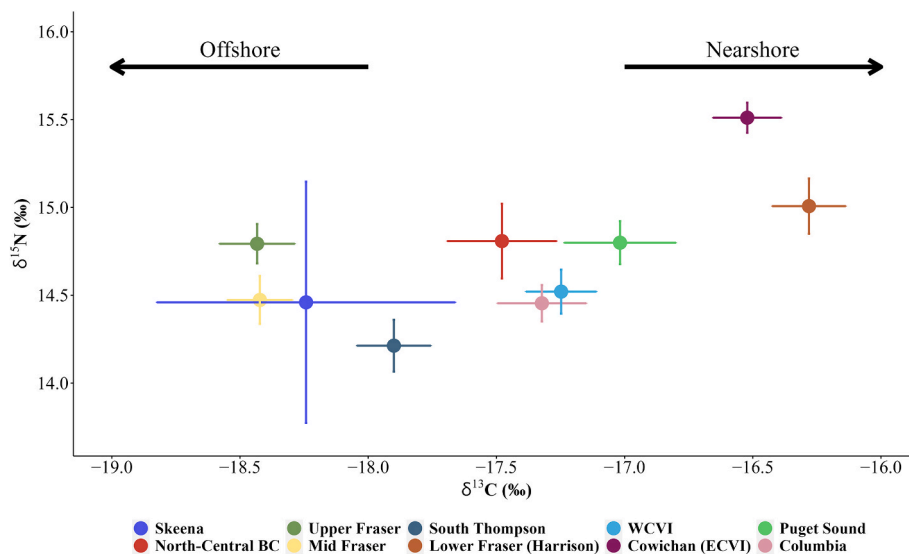


Fig. 2. $\delta^{15}\text{N}$ and lipid normalized $\delta^{13}\text{C}$ (mean \pm SE) for 10 priority Chinook stocks consumed by Resident killer whales. Higher $\delta^{13}\text{C}$ in shelf resident Chinook reflect their use of nearshore marine rearing habitats.

rearing distributions along the continental shelf for the Columbia, WCVI, and North-Central BC Chinook compared to the far north and offshore distribution of Upper and Mid Fraser Chinook (Fig. 1) (Freshwater et al., 2021; Larson et al., 2013; CTC, 2019; Sharma and Quinn, 2012; Shelton et al., 2019; Weitkamp, 2010). The differences observed in mean $\delta^{13}\text{C}$ among the 10 Chinook stocks support previous migration studies that indicate a mosaic of Chinook marine rearing strategies along the continental shelf, the Gulf of Alaska, and offshore habitats (Shelton et al., 2019; Weitkamp, 2010). Mean $\delta^{13}\text{C}$ did not differ between males and females ($p = 0.1$).

Cowichan Chinook mean $\delta^{15}\text{N}$ were higher than those of Columbia, Upper Fraser, Mid Fraser, Puget Sound, Skeena, South Thompson, and West Coast Vancouver Island Chinook (ANOVA; $\delta^{15}\text{N}$ $F_{9,102} = 6.17$, $p < 0.001$, Fig. 2; Tukey post hoc Table S4). Harrison Chinook $\delta^{15}\text{N}$ were higher than South Thompson ($p < 0.001$). These results suggest that Cowichan Chinook and Harrison Chinook are feeding at a higher trophic position, which could contribute to increased concentrations in these particular stocks, relative to the other Chinook stocks (Fig. 2). Mean $\delta^{15}\text{N}$ was higher in males (14.8 ± 0.7) than females (14.6 ± 0.6) ($p = 0.02$), however all Cowichan Chinook were male and therefore could be influencing this difference. These findings should be interpreted with some caution due to the lack of baseline $\delta^{15}\text{N}$ data that could enable a correction for underlying variation in the nitrogen resources supporting different stocks.

3.2. PCB and PBDE concentrations in Chinook stocks

Mean concentrations of $\sum\text{PCBs}$ in all Chinook samples ($n = 83$) ranged from 43.25 ± 1.13 ng/g lw (Upper Fraser) to 825.0 ± 186.7 ng/g lw (Cowichan) with an overall mean $\sum\text{PCBs}$ of 226.0 ± 42.2 ng/g lw. Mean concentrations of $\sum\text{PBDEs}$ in all individuals ($n = 83$) ranged from 2.82 ± 0.43 ng/g lw (Mid Fraser) to 234.4 ± 59.5 ng/g lw (Cowichan) with an overall mean $\sum\text{PBDEs}$ of 53.13 ± 12.89 ng/g lw (Table S8).

$\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations varied among Chinook stocks ($\sum\text{PCBs}$: $\chi^2_{(9)} = 49.6$, $p < 0.001$; $\sum\text{PBDEs}$: $\chi^2_{(9)} = 52.3$, $p < 0.001$) (Fig. 3). Shelf residents (Cowichan and Harrison) had significantly higher ($p < 0.05$) $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations compared to all other stocks, while Harrison $\sum\text{PCB}$ concentrations appeared to be higher than Skeena ($p = 0.054$) (Tables S6 and S7). A greater sample size for Skeena Chinook ($n = 4$) may strengthen this observation. Puget Sound Chinook had higher $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations than Skeena ($\sum\text{PBDEs}$, $p = 0.03$; $\sum\text{PCBs}$, $p = 0.05$), North-Central BC, Mid Fraser, and Upper Fraser Chinook (Tables S6 and S7). No other differences ($p > 0.05$) were found among concentrations of $\sum\text{PCBs}$ and

$\sum\text{PBDEs}$ in the Chinook stocks. Higher $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations in the shelf resident Chinook stocks (Harrison, Cowichan, and Puget Sound) compared to the more offshore migrating stocks likely reflects their proximity to urban centers. For instance, PCB and PBDE concentrations in harbor seals and PBDE concentrations in Dungeness crab (*Cancer magister*) in the coastal waters of British Columbia and the US were highest near urban sites compared to more remote sites (Ikonomou et al., 2006; Ross et al., 2004, 2013).

Harrison Chinook (4.1 ± 0.1 years old; Table 1) analyzed in this study had mean $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations 1.5-fold and 2.2-fold lower than 2.5 year old Harrison Chinook reported in 2009 (Cullon et al., 2009), respectively. Mean Skeena Chinook muscle $\sum\text{PCB}$ concentrations in the present study (Table S8) were similar to whole body adult Skeena Chinook concentrations collected in 2004; however, mean Skeena Chinook $\sum\text{PBDE}$ concentrations in the present study were 5-fold higher than the 2004 adult Skeena $\sum\text{PBDE}$ concentrations (O'Neill et al., 2006). Mean $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations measured in 4 year old South Thompson Chinook from this study were 1.5- and 2.8-fold lower than 4 or 5 year old Thompson (Conservation Unit not specified) Chinook reported in 2010 (Veldhoen et al., 2010), respectively. Mean $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations in Columbia Chinook in this study were approximately 2.5-fold and 2.7-fold lower than levels measured in Columbia Chinook reported in 2006 (Stone, 2006), respectively. Similarly, $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations in Puget Sound Chinook from this study were 10- and 4-fold lower than Puget Sound Chinook concentrations reported in 2009, respectively (Cullon et al., 2009; O'Neill and West, 2009). The Puget Sound Chinook in this study were collected along southwest Vancouver Island or northeast Vancouver Island, whereas those Puget Sound Chinook with higher PCBs and PBDEs reported in 2009 were collected either in river or within Puget Sound. The latter group with the much higher PCB and PBDE levels may have had a significant portion of resident Puget Sound Chinook (locally called 'blackmouth'), which remain within Puget Sound during their marine rearing (Chamberlin et al., 2011; Cullon et al., 2009; O'Neill and West, 2009). Whereas the Puget Sound Chinook collected from our study are likely fish that resided in Puget Sound for several months as juveniles then migrated out of Puget Sound to rear along the coast of the Pacific Ocean and therefore have a different marine life history than the residents (Duffy et al., 2005; Simenstad et al., 1982). The decreases in $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations among most of these stocks over time mirror temporal trends of declining concentrations in Pacific herring (*Clupea pallasii*), English sole (*Parophrys vetulus*), and harbor seals from Puget Sound, and seabirds from the northeast Pacific (Elliott et al., 2023; Ross et al., 2013; West et al., 2017).

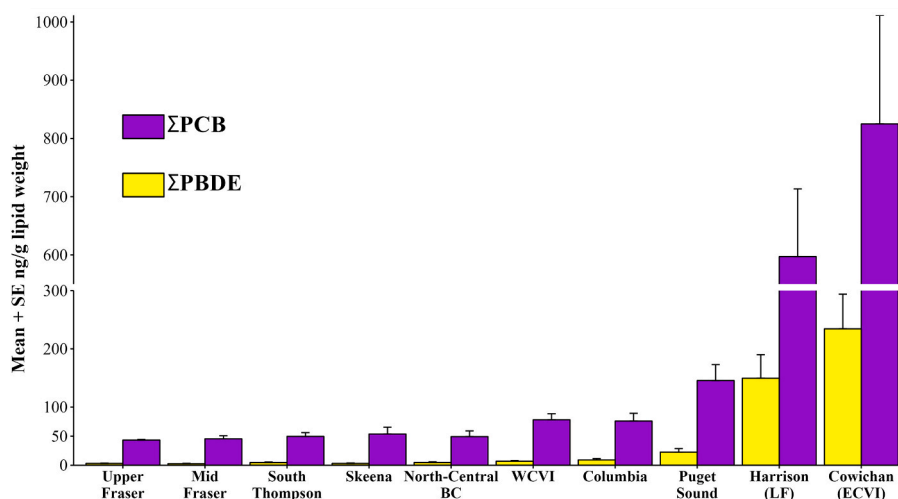


Fig. 3. Mean $\sum\text{PCB}$ and $\sum\text{PBDE}$ concentrations in 10 priority Chinook stocks, with PCBs found at 3.5 (Cowichan) to 17 (Skeena) -fold higher than PBDEs. Error bars are standard error. Stocks are ordered from farthest to closest in proximity to land-based pollution.

The top six PCB congeners, which included a combination of PCB 52, 70, 99, 101, 118, 138, 149 and 153, accounted for $34.9 \pm 0.2\%$ to $44.3 \pm 1.1\%$ of the \sum PCBs among all Chinook (Tables S8 and S10). PCB 153 ($8.04 \pm 0.14\%$ to $13.0 \pm 0.6\%$ of the total PCBs), PCB 138 ($6.47 \pm 0.24\%$ to $10.1 \pm 0.5\%$), and PCB 101 ($5.68 \pm 0.09\%$ to $6.62 \pm 0.26\%$) were consistently the top three congeners among all Chinook stocks. These results were similar to the top PCB congeners in Pacific salmon transplanted to the Great Lakes, masu salmon (*Oncorhynchus masou masou* and *Oncorhynchus masou ishikawae*) from Japan, and Chinook salmon previously sampled within the Salish Sea (Gerig et al., 2016; Matsumoto et al., 2014; Pearce, 2018). These three PCB congeners are known to biomagnify in marine food webs and are commonly reported in upper trophic level marine mammals, such as harbour porpoises, harbour seals, and RWKs (Madgett et al., 2022; Pearce, 2018).

The fourth, fifth and sixth top PCB congeners in the shelf resident Chinook (Harrison, Cowichan, and Puget Sound) were dominated by heavier and more chlorinated PCBs (149, 99, and 118) compared to the offshore and far north rearing Chinook (Upper Fraser, Mid Fraser, North-Central BC) which tended to be lighter (PCB 70, PCB 52) (Tables S8 and S11). PCB 70 was the fourth (Upper and Mid Fraser) or sixth (Skeena, North-Central BC, South Thompson) top congener in five stocks with relatively lower \sum PCBs and that rear in more offshore areas (Tables S8 and S11). PCB 52 was in the top six congeners in the three stocks (North-Central BC, Upper, and Mid Fraser) with the lowest \sum PCBs and more offshore marine rearing (Tables S8 and S11).

The top six PBDE congeners, which included a combination of 28/33, 47, 49, 99, 100, 154, and 209, together accounted for $83.7 \pm 1.1\%$ to $94.1 \pm 1.1\%$ of the total PBDE in all Chinook (Tables S8 and S11). PBDE 47 was consistently the top PBDE congener among all Chinook stocks, accounting for $46.6 \pm 6.3\%$ to $61.0 \pm 1.1\%$ of \sum PBDE (Tables S8 and S11). PBDE 99 ($7.65 \pm 0.61\%$ to $12.2 \pm 1.0\%$) and 100 ($8.11 \pm 1.13\%$ to $10.8 \pm 0.4\%$) were the second and third most abundant PBDE congeners in eight of the 10 stocks (Tables S8 and S11). PBDE 100 was the second-most dominant congener among the shelf residents (Cowichan, Harrison, Puget Sound), whereas PBDE 99 was the second-most dominant congener among the offshore stocks (Columbia, Mid Fraser, North-Central BC, South Thompson) (Tables S8 and S11). In wild Alaskan Chinook salmon and Pacific salmon transplanted to the Great Lakes, PBDE 47 was also the dominant congener with PBDE 99 and 100 accounting for the second and/or third highest percent contribution to \sum PBDE (Gerig et al., 2016; Shaw et al., 2008). PBDE 49 was the fourth top congener for nine of the 10 stocks ($6.49 \pm 0.58\%$ to $9.07 \pm 0.64\%$; Tables S8 and S11). PBDE 209 was detected in all but one of the stocks (WCVI) and was the fifth top congener in five of the more offshore Chinook stocks (Table S8) but was not one of the dominant congeners in the shelf rearing resident stocks. This is surprising, as PBDE 209 is generally associated with more urbanized areas (Gewurtz et al., 2011). The top ranking placement for the offshore Chinook stocks can be explained by the mean total PBDE concentrations in offshore stocks being 83-fold lower than shelf residents. These findings and the overall detection of PBDE-209 in the present study are likely a result of a laboratory contamination issue, which is a commonly reported problem for this particular congener (Alcock et al., 2011; Gewurtz et al., 2011).

3.3. Multivariate analysis of PCBs and PBDEs in 10 stocks of adult Chinook

After treating the PCB congener data, 113 congeners were used in the final PCA. The first principal component (p1 & t1: 63.6%) for PCB congeners differentiated Chinook salmon with a greater proportion of lighter congeners from Chinook with a greater proportion of heavier congeners (Fig. 4A & C). Polychlorinated biphenyl congeners with a degree of chlorination ≤ 5 (lighter) projected on the left of p1 while congeners with a degree of chlorination > 5 (heavier) projected on the right of p1 (Fig. 4C). A light PCB signature is characteristic of long-range atmospheric or oceanic signal, which is the result of more volatile

(lighter) congeners travelling greater distances, whereas a heavier signature reflects a nearby source signal, dominated by heavier more chlorinated congeners (Brown et al., 2014; Ross et al., 2004).

Shelf resident (Cowichan and Harrison) and Puget Sound Chinook projected to the right of the PCA (Fig. 4A) indicating that these stocks had a greater proportion of heavier congeners (Fig. S1), consistent with the individuals from these stocks utilizing more coastal habitats (Fig. 2) and closer proximity to land-based pollution sources (Burd et al., 2022). The offshore Chinook (Mid and Upper Fraser) projected to the left of the PCA score plot (Fig. 4A) indicating that they were dominated by a higher proportion of lighter congeners (chlorination < 5 ; Fig. S1), which is consistent with distant PCB sources and reflects their utilization of more offshore habitats (depleted $\delta^{13}\text{C}$ signature; Fig. 2) that are further from regional urban sources (Brown et al., 2014; Yunker et al., 2011). South Thompson, Skeena, North-Central BC, and WCVI also projected on the left side of t1 but to a lesser extent than the offshore stocks, following their known marine distribution patterns to the far north in the Gulf of Alaska or to southeast Alaska (WCVI) compared with the Upper and Mid Fraser Chinook, which migrate further offshore. Columbia Chinook did not project to one side or the other but were spread across t1.

These divergent PCB signatures among the Chinook stocks are consistent with observations in other species, including other species of salmon, in which proximity to regional or local sources explained the heavier PCB profiles compared with those in more remote locations (Brown et al., 2014; Cullon et al., 2009; Ross et al., 2004; Yunker et al., 2011). In addition, similar results have been observed in wild coho salmon PCB signatures that separated coho into “coastal” and “remote” groups (Yunker et al., 2011). The “remote” coho grouped with wild chum, sockeye, and pink salmon, known to have extensive offshore marine distributions, while the “coastal” coho grouped with wild Chinook with marine distributions closer to the continental shelf (Yunker et al., 2011). Japanese masu salmon inhabiting waters closer to an industrial area showed heavier PCB profiles and more enriched $\delta^{13}\text{C}$ compared to masu salmon in waters farther away from an industrial area (Matsumoto et al., 2014).

The log concentrations of \sum PCBs in Chinook tissue samples positively correlated with t1 scores ($r^2 = 0.75$; $p < 0.001$; Fig. S3A). This is consistent with Fig. 4 results that show shelf resident Chinook having heavier PCB signatures and higher \sum PCB concentrations (Table S8) relative to the far north and offshore distributed Chinook with lighter PCB signatures. The log K_{ow} and p1 (the variable loadings of the first principal component of the individual PCB congeners) also correlated ($r^2 = 0.706$; $p < 0.001$). These results are comparable to those of Brown et al. (2014) showing a PCB profile that strongly correlated with both \sum PCB concentrations and log K_{ow} (octanol-water partition coefficient which describes the partitioning between lipid and water).

Out of a total of 40 PBDE congeners, 14 were used in the final PBDE PCA. The first (p1) and second (p2) principal components explained 47.3% and 15.11% of the variability in the PBDE congener patterns, respectively (62.44% total; Fig. 4B & D). The first principal component (p1: 47.3%) for PBDE congeners separated based on their biomagnification and bioaccumulation potential in Chinook tissues. For example, PBDE 47 (tetra) and PBDE 100 (penta), which projected on the left side of p1, are highly lipophilic, bioaccumulative (log $K_{ow} > 6.5$) congeners that Chinook salmon and other fish species cannot readily metabolize or debrominate resulting in greater accumulation of these congeners in their tissues over time (Roberts et al., 2011; Shaw et al., 2008). Further, PBDE-99 and -100 have been found to biotransform to PBDE 47 through debromination in the food web (Kelly et al., 2008; Lv et al., 2020). This would contribute to the higher contribution of PBDE 47 to total PBDEs over time in higher trophic level species like Chinook.

In contrast, the PBDE congeners projected on the right side of p1 were a mix of di, tri, tetra, penta, and hexa congeners (e.g., 15, 17/25, 66, 119/120, 155), but are not known to accumulate to high concentrations in Chinook tissues (Shaw et al., 2008). The separation of PBDE congeners along the second principal component (p2: 15.11%) may be

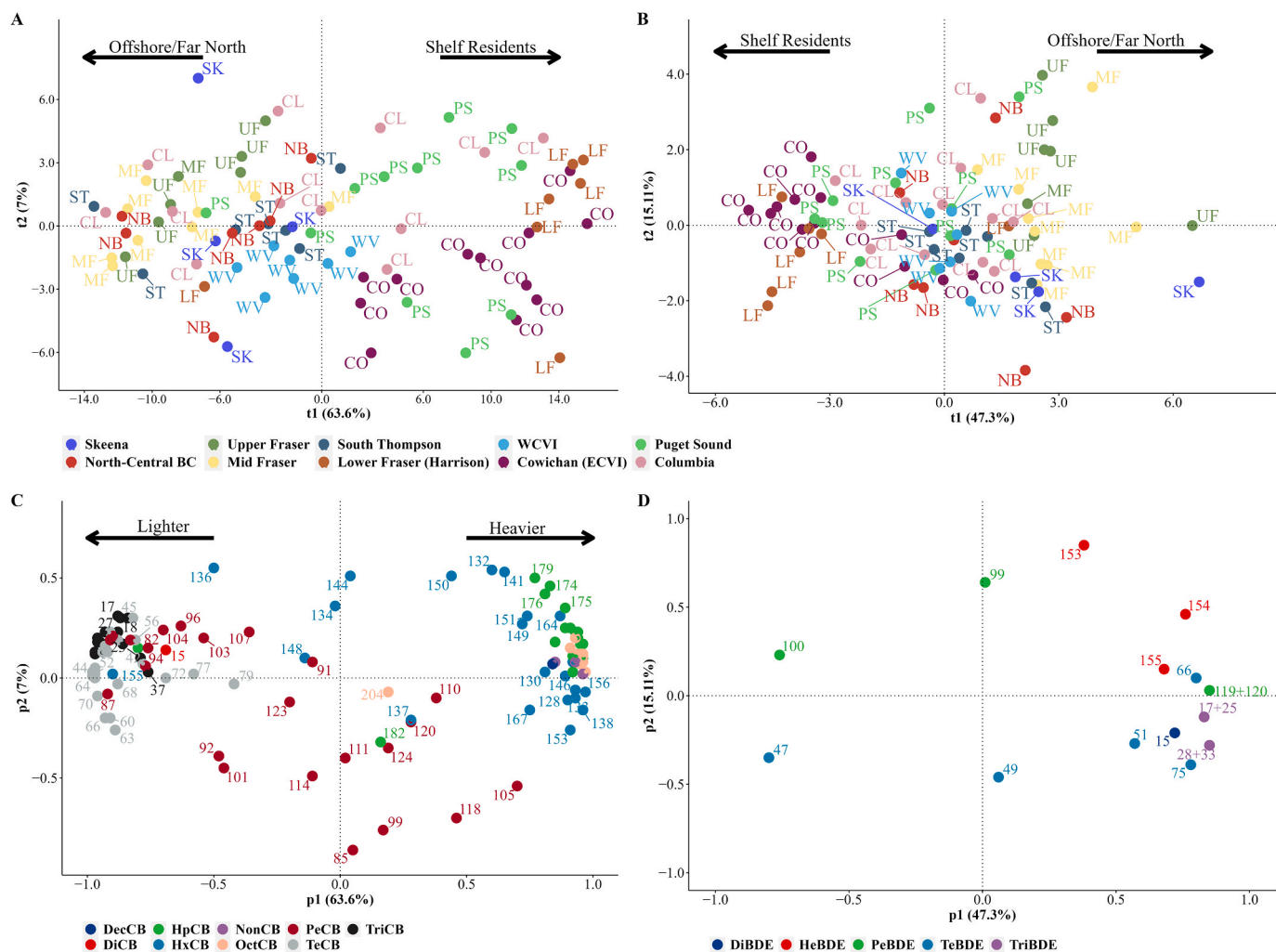


Fig. 4. Principal components analysis (PCA) of PCB (A, C) and PBDE (B, D) congeners in 10 stocks of adult Chinook salmon. Chinook samples (A, B) are color coded by stock, and PCB and PBDE congeners (C, D) are color coded by homologue (degree of chlorination or bromination) group. PCB homologue abbreviations: DiCB (dichlorobiphenyl), TriCB (trichloro), TeCB (tetrachloro), PeCB (pentachloro), HxCB (hexachloro), HpCB (heptachloro), OctCB (octachloro), NonCB (nonachloro), DecCB (decachloro). PBDE homologue abbreviations: DiBDE (dibromodiphenyl ether), TriBDE (tribromo), TeBDE (tetrabromo), PeBDE (pentabromo), HeBDE (hexabromo).

shaped largely by metabolism. Congeners 153 and 99 projected on the top of p2 have the potential to undergo metabolic debromination in fish species, specifically Chinook for PBDE-99 to -66 and -49 (Roberts et al., 2011). However, congeners -47, -49, -51, and -75 which projected on the bottom of p2 are not readily debrominated (Dietrich et al., 2015; Ng et al., 2018). The log K_{OW} did not correlate with p1 ($p > 0.05$) but did correlate with p2 scores ($r^2 = 0.622$; $p = 0.002$).

The Chinook PBDE scores plot along t1 (47.3%) also separated based on stock and associated marine distributions. The shelf residents (Cowichan and Harrison) projected closely together on the left side of t1, and offshore Chinook (Upper and Middle Fraser) projected on the right side of t1. The log concentration of \sum PBDEs in Chinook tissue negatively correlated with the t1 scores of Chinook samples ($r^2 = 0.79$; $p < 0.001$; Fig. S3B), which is consistent with our results above that showed bioaccumulative PBDE congeners were more dominant in shelf resident Chinook with higher PBDE concentrations (Table S8) relative to the far north or offshore distributed stocks.

Feeding ecology helped explain the PCB and PBDE patterns observed among the 10 Chinook stocks. The PCB t1 scores (the sample scores of the first principal component) for Chinook were positively correlated with $\delta^{13}C$ and $\delta^{15}N$ and negatively correlated with percent lipid (Fig. 5A and B; Fig. 6A). Although there are strong relationships between the PCB

t1 variability and the individual biological variables, the interaction between $\delta^{13}C$ and stock best explained the variability in the PCB t1 scores ($F_{10,72} = 15.81$, $r^2 = 0.644$, $p < 0.001$; Table 2), indicating Chinook stock PCB congener patterns are largely shaped by coastal versus offshore use of carbon resources. Similarly, PBDE t1 scores were negatively correlated with $\delta^{13}C$ and $\delta^{15}N$ (Fig. 5C and D) and positively correlated with percent lipid (Fig. 6B). However, the model that best explained the variation in PBDE t1 scores included $\delta^{13}C$, $\delta^{15}N$, percent lipid, and the interaction between $\delta^{15}N$ and percent lipid (Table 2). Trophic level and metabolism (lipid content), as well as carbon food source, seem to be driving PBDE congener patterns in Chinook salmon.

3.4. Cluster analyses of Chinook PCB and PBDE congeners

Applying k-means and hierarchical clustering methods, we separated the Chinook stocks into three clusters, which largely reflected their stock, feeding ecology ($\delta^{13}C$), and marine rearing habitat: Cluster 1 (Cowichan, Harrison, Puget Sound), Cluster 2 (Skeena, North-Central BC, Upper Fraser, Mid Fraser, South Thompson, and WCVI), and Cluster 3 (Columbia) (Fig. 7). Using their marine rearing distributions and stock affiliations, we defined the groupings as: "Shelf residents" for Cluster 1, "Far North/Offshore" for Cluster 2, and "Columbia" for Cluster

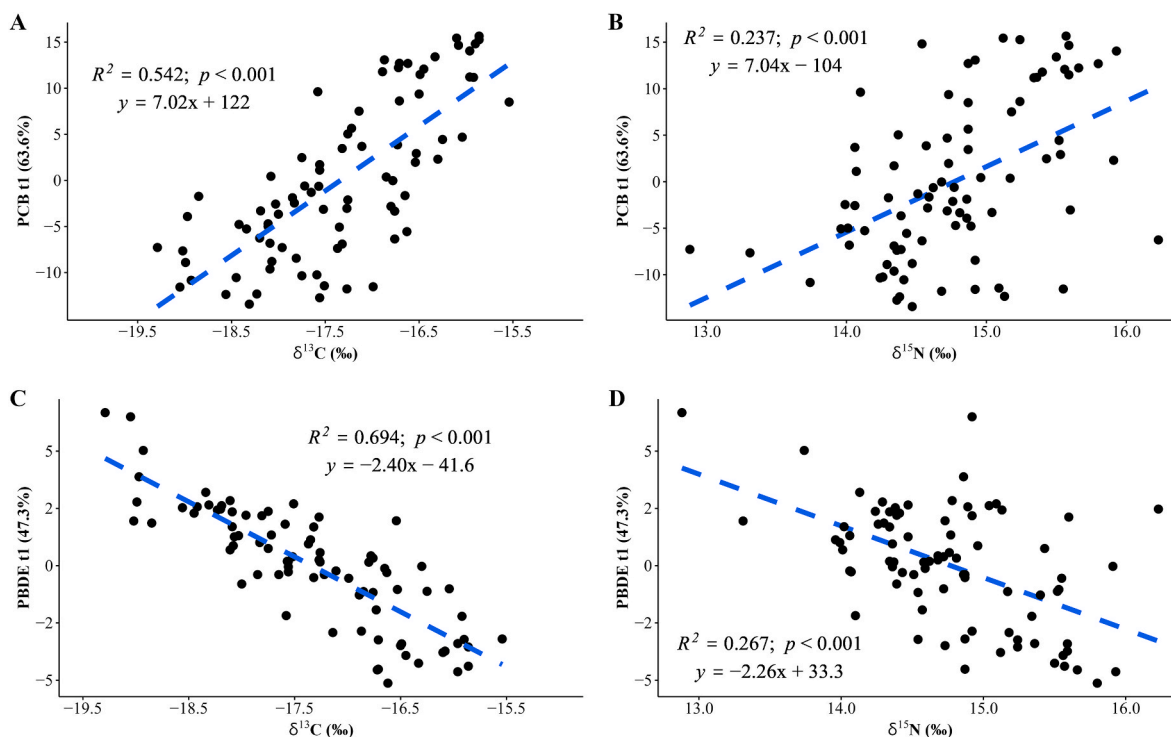


Fig. 5. Chinook salmon PCB t1 and PBDE t1 scores positively and negatively correlated with $\delta^{15}\text{N}$ and lipid normalized $\delta^{13}\text{C}$, respectively.

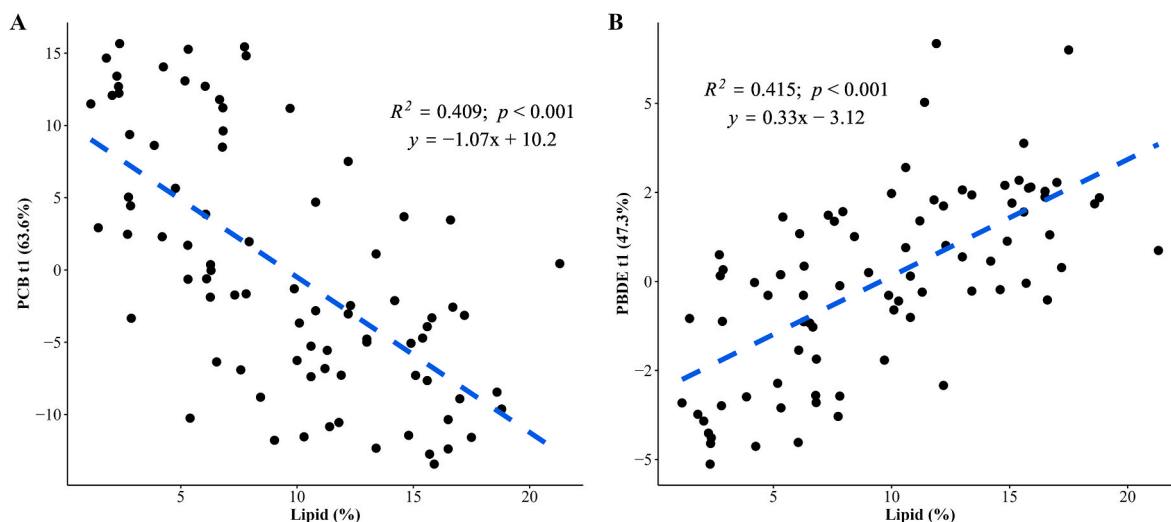


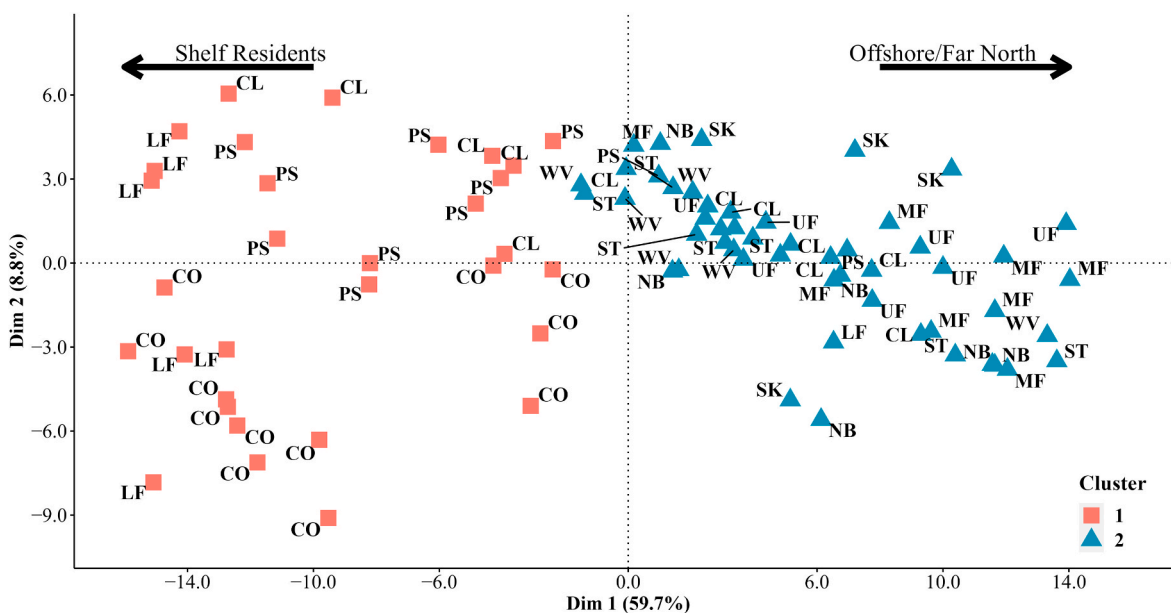
Fig. 6. Chinook salmon PCB t1 and PBDE t1 scores negatively and positively correlated with percent lipid, respectively.

Table 2

Akaike Information Criterion (AICc) assessment for models describing variation in the first principal components (t1) of PCB and PBDE PCAs in combination with backwards stepwise regression.

PCB t1	F	R ² (adjusted)	p value	K	AICc	ΔAICc	w _i
$\delta^{13}\text{C}$ (‰) * Stock	15.81 _{10,72}	0.644	<0.001	12	522.8	0	0.66
$\delta^{13}\text{C}$ (‰) + $\delta^{15}\text{N}$ (‰) + Stock + Stock * $\delta^{13}\text{C}$ (‰)	11.15 _{20,62}	0.712	<0.001	22	525	2.25	0.21
$\delta^{13}\text{C}$ (‰) + $\delta^{15}\text{N}$ (‰) + Lipid + Stock + Stock * $\delta^{13}\text{C}$ (‰)	10.74 _{21,61}	0.714	<0.001	23	527.1	4.31	0.08
PBDE t1	F	R² (adjusted)	p value	K	AICc	ΔAICc	w_i
$\delta^{13}\text{C}$ (‰) + $\delta^{15}\text{N}$ (‰) + Lipid + $\delta^{15}\text{N}$ (‰) * Lipid	55.93 _{4,78}	0.728	<0.001	6	293.3	0	0.95
$\delta^{13}\text{C}$ (‰)	186.7 _{1,81}	0.694	<0.001	3	299.6	6.26	0.04
$\delta^{13}\text{C}$ (‰) * Stock	22.42 _{10,72}	0.723	<0.001	12	303.5	10.22	0.01

The model chosen by backwards stepwise regression is indicated in bold.



	Skeneea	North-Central BC	Upper Fraser	Mid Fraser	South Thompson	Lower Fraser (Harrison)	WCVI	Cowichan (ECVI)	Puget Sound	Columbia
Cluster 1	0	0	0	0	0	6	0	12	9	5
Cluster 2	4	7	7	9	7	1	7	0	2	7

Fig. 7. K-means clustering of PCB and PBDE congener patterns in ten Chinook stocks. Points are labeled by abbreviated stock name. SK—Skeneea, NB—North-Central BC, UF—Upper Fraser, MF—Mid Fraser, ST—South Thompson, LF—Lower Fraser (Harrison), WV—West Coast Vancouver Island, CO—Cowichan, PS—Puget Sound, CL—Columbia.

3. Individual k-means and hierarchical cluster analyses showed similar results to the combined k-means clustering (Fig. 7; Fig. S4). To help us determine these clusters we set a criterion, wherein, at least 80% of individuals from an identified Chinook stock had to be grouped into one cluster in order for all individuals from that stock to be placed into a defined cluster. For example, nine out of eleven individuals (82%) from Puget Sound were in Cluster 1, and therefore placed in Cluster 1 “Shelf residents.” Similar to the PCA results above (Fig. 4A & B), the Columbia stock had a distinct pattern from the other two clusters, distributing almost evenly between Cluster 1 ($n = 5$) and 2 ($n = 7$) (Fig. 7), and thus was placed in its own defined cluster. Further investigation of the different Columbia populations that make up our defined Columbia stock is warranted. Such examination would require a larger dataset to see if variations in feeding ecology and or contaminant profiles can pick up their varied marine distributions. These clusters can be used to simplify future analyses when evaluating differences of other contaminant classes in these Chinook stocks.

4. Conclusions

The evaluation of PCB and PBDE concentrations and congener patterns and stable isotope profiles in these 10 Chinook salmon stocks sheds light on the role that feeding ecology and distribution in marine rearing grounds play in shaping the accumulation of these two contaminants of concern. We conclude that shelf resident Chinook stocks that feed closer to the coast consume prey that is more contaminated because of proximity to land-based sources of urban and industrial pollution. These rearing grounds include Puget Sound and the Strait of Georgia within the Salish Sea. In contrast, Chinook that feed further offshore consume prey contaminated by more ‘global’ or ‘long-range’ sources, with lower levels of POPs. With the offshore upper and middle Fraser Chinook stocks being typically more lipid-rich, they are a critical component of the diet of SRKW (Hanson et al., 2010). Given the dramatic decline of these stocks, especially in the last decade (Atlas et al., 2023; COSEWIC, 2018), SRKW may be compelled to progressively shift their prey base to lipid-poor shelf resident Chinook stocks that have higher contaminant

levels (Cullon et al., 2009; Hanson et al., 2021). Efforts to increase the abundance of offshore upper and middle Fraser Chinook stocks should be prioritized, as well as accelerate the virtual elimination of PCB and PBDE sources and hotspots in coastal waters. This approach would have broad benefit for Chinook salmon and *Endangered* SRKW.

Credit author contributions

Stephanie Holbert: Methodology, Investigation, Data curation, Formal analysis, Visualization, Writing – original draft, writing—reviewing and editing. Katerina Colbourne: Methodology, Writing – review & editing. Aaron Fisk: Methodology; writing—reviewing and editing. Peter Ross: Methodology; writing—reviewing and editing. Misty MacDuffee: writing—reviewing and editing; Frank Gobas: writing—reviewing and editing. Tanya Brown: Conceptualization, Investigation, Methodology, Data curation, Resources, Funding acquisition, Supervision, writing—reviewing and editing.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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References

- Aitchison, J., 1986. *The Statistical Analysis of Compositional Data*.
- Alcock, R.E., MacGillivray, B.H., Busby, J.S., 2011. Understanding the mismatch between the demands of risk assessment and practice of scientists—the case of Deca-BDE. *Environ. Int.* 37 (1), 216–225. <https://doi.org/10.1016/j.envint.2010.06.002>.
- Atlas, W.L., Sloat, M.R., Satterthwaite, W.H., Buehrens, Thomas W., Parken, C.K., Moore, J.W., Mantua, Nathan J., Hart, J., Potapova, A., 2023. Trends in Chinook salmon spawner abundance and total run size highlight linkages between life history, geography and decline. *Fish Fish.* 24 (4), 595–617. <https://doi.org/10.1111/faf.12750>.
- Beacham, T.D., Candy, J.R., Jonsen, K.L., Supernault, J., Wetklo, M., Deng, L., Miller, K. M., Withler, R.E., Varnavskaya, N., 2006. Estimation of stock composition and individual identification of chinook salmon across the Pacific Rim by use of microsatellite variation. *Trans. Am. Fish. Soc.* 135 (4), 861–888. <https://doi.org/10.1577/t05-241.1>.
- Beacham, T.D., Wallace, C., MacConnachie, C., Jonsen, K., McIntosh, B., Candy, J.R., Withler, R.E., 2018. Population and individual identification of Chinook salmon in British Columbia through parentage-based tagging and genetic stock identification with single nucleotide polymorphisms. *Can. J. Fish. Aquat. Sci.* 75 (7), 1096–1105. <https://doi.org/10.1139/cjfas-2017-0168>.
- Beamish, R., Lange, K., Neville, C., Sweeting, R., Beacham, T., 2011. Structural patterns in the distribution of ocean- and stream-type juvenile chinook salmon populations in the Strait of Georgia in 2010 during the critical early marine period. *NPAFC Doc* 1354, 27.
- Brown, G., Baillie, S., Thiess, M., Bailey, R., Candy, J., Parken, C., Willis, D., 2019. *Pre-COSEWIC review of southern British Columbia Chinook Salmon (*Oncorhynchus tshawytscha*) conservation units, Part I: background*. DFO Can. Sci. Adv. Sec. Res. Doc 11, 2019.
- Brown, T.M., Fisk, A.T., Helbing, C.C., Reimer, K.J., 2014. Polychlorinated biphenyl profiles in ringed seals (*Pusa hispida*) reveal historical contamination by a military radar station in Labrador, Canada. *Environ. Toxicol. Chem.* 33 (3), 592–601. <https://doi.org/10.1002/etc.2468>.
- Burd, B.J., Lowe, C.J., Morales-Caselles, C., 2022. Uptake of PCBs into sediment dwellers and trophic transfer in relation to sediment conditions in the Salish Sea. *FACTETS*. <https://doi.org/10.1139/facets-2021-0032>.
- Burnham, K.P.A., David, R., 2002. *Model Selection and Multimodel Inference: a Practical Information-Theoretic Approach*, second ed. Springer-Verlag.
- Chamberlin, J.W., Essington, T.E., Ferguson, J.W., Quinn, T.P., 2011. The influence of hatchery rearing practices on salmon migratory behavior: is the tendency of Chinook salmon to remain within Puget Sound affected by size and date of release? *Trans. Am. Fish. Soc.* 140 (5), 1398–1408. <https://doi.org/10.1080/00028487.2011.623993>.
- Christensen, J.R., MacDuffee, M., Macdonald, R.W., Whitticar, M., Ross, P.S., 2005. Persistent organic pollutants in British Columbia grizzly bears: consequence of divergent diets. *Environ. Sci. Technol.* 39 (18), 6952–6960. <https://doi.org/10.1021/es050749f>.
- COSEWIC, 2018. *COSEWIC Assessment and Status Report on the Chinook Salmon *Oncorhynchus tshawytscha* Designatable Units in Southern British Columbia (Part One – Designatable Units with No or Low Levels of Artificial Releases in the Last 12 Years)*. Canada. Committee on the Status of Endangered Wildlife in Canada, Ottawa, Canada xxxi + 283 pp. <https://www.canada.ca/en/environment-climate-change/services/species-risk-public-registry/cosewic-assessments-status-reports/chinook-salmon-2018.html>.
- Cullon, D.L., Yunker, M.B., Alleyne, C., Dangerfield, N.J., O'Neill, S., Whitticar, M.J., Ross, P.S., 2009. Persistent organic pollutants in Chinook salmon (*Oncorhynchus tshawytscha*): implications for resident killer whales of British Columbia and adjacent waters. *Environ. Toxicol. Chem.: Int. J.* 28 (1), 148–161. <https://doi.org/10.1897/08-125.1>.
- CTC (Chinook salmon Technical Committee). 2019. *Pacific Salmon Commission Joint Chinook salmon Technical Committee Report. Annual report of catch and escapement for 2018*. Report TCChinook (19)-01. Pacific Salmon Commission, Vancouver, BC, Canada. 241 p.
- Das, K., Holsbeek, L., Browning, J., Siebert, U., Birkun Jr., A., Bouquegneau, J.-M., 2004. Trace metal and stable isotope measurements ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) in the harbour porpoise *Phocoena phocoena relicta* from the Black Sea. *Environ. Pollut.* 131 (2), 197–204. <https://doi.org/10.1016/j.envpol.2004.02.006>.
- Di Paolo, C., Gandhi, N., Bhavsar, S.P., Van den Heuvel-Greve, M., Koelmans, A.A., 2010. Black carbon inclusive multichemical modeling of PBDE and PCB biomagnification and transformation in estuarine food webs. *Environ. Sci. Technol.* 44 (19), 7548–7554. <https://doi.org/10.1021/es101247e>.
- Dietrich, J.P., Strickland, S.A., Hutchinson, G.P., Van Gaest, A.L., Krupnik, A.B., Ylitalo, G.M., Arkoosh, M.R., 2015. Assimilation efficiency of PBDE congeners in Chinook salmon. *Environ. Sci. Technol.* 49 (6), 3878–3886. <https://doi.org/10.1021/es5057038>.
- Doutaz, D., Weir, L., Arbeider, M., Braun, D., Jenewein, B., Rickards, K., Labelle, M., Curtis, S., Mozin, P., Whitney, C., Parken, C., Bailey, R., 2021. *Recovery Potential Assessment for 11 Designatable Units of Fraser River Chinook Salmon, *Oncorhynchus tshawytscha*, Part 1: Elements 1 to 11*, pp. xiii–337, 2021/06/3.
- Duffy, E.J., Beauchamp, D.A., Buckley, R.M., 2005. Early marine life history of juvenile Pacific salmon in two regions of Puget Sound. *Estuar. Coast Shelf Sci.* 64 (1), 94–107. <https://doi.org/10.1016/j.ecss.2005.02.009>.
- Elliott, J.E., Kesic, R., Lee, S.L., Elliott, K.H., 2023. Temporal trends (1968–2019) of legacy persistent organic pollutants (POPs) in seabird eggs from the northeast Pacific: is it finally twilight for old POPs? *Sci. Total Environ.* 858, 160084 <https://doi.org/10.1016/j.scitotenv.2022.160084>.
- Environment and Climate Change Canada, 2020. *Southern Resident Killer Whale Contaminants Technical Working Group Accomplishment Highlights and Recommendations*, p. 2020. https://publications.gc.ca/collections/collection_2020/eccc/En4-416-2020-eng.pdf.
- Erickson, M.D., 1997. *Analytical Chemistry of PCBs*, second ed. Lewis Publishers, CRC Press.
- Fisk, A.T., Hobson, K.A., Norstrom, R.J., 2001. Influence of chemical and biological factors on trophic transfer of persistent organic pollutants in the Northwest Polynya marine food web. *Environ. Sci. Technol.* 35 (4), 732–738. <https://doi.org/10.1021/es001459w>.
- Ford, J.K.B., Ellis, G.M., 2006. Selective foraging by fish-eating killer whales *Orcinus orca* in British Columbia. *Mar. Ecol. Prog. Ser.* 316, 185–199. <https://doi.org/10.3354/meps316185>.
- Ford, J.K.B., Wright, B.M., Ellis, G.M., Candy, J.R., 2010. *Chinook Salmon Predation by Resident Killer Whales: Seasonal and Regional Selectivity, Stock Identity of Prey, and Consumption Rates*. DFO Canadian Science Advisory Secretariat, pp. iv–43. *Doc.* 2009/101.
- Ford, M.J., 2022. *Biological Viability Assessment Update for Pacific Salmon and Steelhead Listed under the Endangered Species Act: Pacific Northwest*.
- Freshwater, C., Anderson, S.C., Beacham, T.D., Luedke, W., Wor, C., King, J., 2021. An integrated model of seasonal changes in stock composition and abundance with an application to Chinook salmon. *PeerJ* 9, e11163. <https://doi.org/10.7717/peerj.11163>.
- Gerig, B.S., Chaloner, D.T., Janetski, D.J., Rediske, R.R., O'Keefe, J.P., Moerke, A.H., Lamberti, G.A., 2016. Congener patterns of persistent organic pollutants establish the extent of contaminant biotransport by Pacific salmon in the Great Lakes. *Environ. Sci. Technol.* 50 (2), 554–563. <https://doi.org/10.1021/acs.est.5b05091>.
- Gewurtz, S.B., McGoldrick, D.J., Clark, M.G., Keir, M.J., Malecki, M.M., Gledhill, M., Sekela, M., Syrgiannis, J., Evans, M.S., Armellin, A., 2011. Spatial trends of polybrominated diphenyl ethers in Canadian fish and implications for long-term monitoring. *Environ. Toxicol. Chem.* 30 (7), 1564–1575. <https://doi.org/10.1002/etc.555>.
- Gustafson, R.G., Waples, R.S., Myers, J.M., Weitkamp, L.A., Bryant, G.J., Johnson, O.W., Hard, J.J., 2007. Pacific salmon extinctions: quantifying lost and remaining diversity. *Conserv. Biol.* 21 (4), 1009–1020. <https://doi.org/10.1111/j.1523-1739.2007.00693.x>.
- Hanson, M.B., Baird, R.W., Ford, J.K.B., Hempelmann-Halos, J., Van Doornik, D.M., Candy, J.R., Emmons, C.K., Schorr, G.S., Gisborne, B., Ayres, K.L., Wasser, S.K., Balcomb, K.C., Balcomb-Bartok, K., Sneva, J.G., Ford, M.J., 2010. Species and stock identification of prey consumed by endangered southern resident killer whales in their summer range. *Endanger. Species Res.* 11, 69–82. <https://doi.org/10.3354/esr00263>.
- Hanson, M.B., Emmons, C.K., Ford, M.J., Everett, M., Parsons, K., Park, L.K., Hempelmann, J., Van Doornik, D.M., Schorr, G.S., Jacobsen, J.K., Sears, M.F., Sears, M.S., Sneva, J.G., Baird, R.W., Barre, L., 2021. Endangered predators and endangered prey: seasonal diet of Southern Resident killer whales. *PLoS One* 16 (3), e0247031. <https://doi.org/10.1371/journal.pone.0247031>.
- Hearsey, J.W., Kinziger, A.P., 2015. Diversity in sympatric chinook salmon runs: timing, relative fat content and maturation. *Environ. Biol. Fish.* 98, 413–423. <https://doi.org/10.1007/s10641-014-0272-5>.
- Hendry, A.P., Berg, O.K., 1999. Secondary sexual characters, energy use, senescence, and the cost of reproduction in sockeye salmon. *Can. J. Zool.* 77 (11), 1663–1675. <https://doi.org/10.1139/z99-158>.
- Ikonou, M.G., Fernandez, M.P., Hickman, Z.L., 2006. Spatio-temporal and species-specific variation in PBDE levels/patterns in British Columbia's coastal waters. *Environ. Pollut.* 140 (2), 355–363. <https://doi.org/10.1016/j.envpol.2005.06.031>.
- Kelly, B.C., Ikonou, M.G., Blair, J.D., Gobas, F.A., 2008. Bioaccumulation behaviour of polybrominated diphenyl ethers (PBDEs) in a Canadian Arctic marine food web. *Sci. Total Environ.* 401 (1–3), 60–72. <https://doi.org/10.1016/j.scitotenv.2008.03.045>.
- Kiljunen, M., Grey, J., Sinisalo, T., Harrod, C., Immonen, H., Jones, R.I., 2006. A revised model for lipid-normalizing $\delta^{13}\text{C}$ values from aquatic organisms, with implications for isotope mixing models. *J. Appl. Ecol.* 43 (6), 1213–1222. <https://doi.org/10.1111/j.1365-2664.2006.01224.x>.
- Lallas, P.L., 2001. *The Stockholm Convention on persistent organic pollutants*. *Am. J. Int. Law* 95 (3), 692–708.
- Larocque, S.M., Fisk, A.T., Johnson, T.B., 2021. Evaluation of muscle lipid extraction and non-lethal fin tissue use for carbon, nitrogen, and sulfur isotope analyses in adult

- salmonids. *Rapid Commun. Mass Spectrom.* 35 (12), e9093 <https://doi.org/10.1002/rcm.9093>.
- Larson, W.A., Utter, F.M., Myers, K.W., Templin, W.D., Seeb, J.E., Guthrie Iii, C.M., Bugaev, A.V., Seeb, L.W., Moran, P., 2013. Single-nucleotide polymorphisms reveal distribution and migration of chinook salmon (*Oncorhynchus tshawytscha*) in the Bering sea and North Pacific Ocean. *Can. J. Fish. Aquat. Sci.* 70 (1), 128–141. <https://doi.org/10.1139/cjfas-2012-0233>.
- Lerner, J.E., Hunt, B.P., 2023. Seasonal variation in the lipid content of Fraser River Chinook salmon (*Oncorhynchus tshawytscha*) and its implications for southern resident killer whale (*Orcinus orca*) prey quality. *Sci. Rep.* 13 (1), 2675. <https://doi.org/10.1038/s41598-023-28321-9>.
- Lv, M., Tang, X., Zhao, Y., Li, J., Zhang, B., Li, L., Jiang, Y., Zhao, Y., 2020. The toxicity, bioaccumulation and debromination of BDE-47 and BDE-209 in *Chlorella sp.* under multiple exposure modes. *Sci. Total Environ.* 723, 138086. <https://doi.org/10.1016/j.scitotenv.2020.138086>.
- Madgett, A.S., Yates, K., Webster, L., McKenzie, C., Brownlow, A., Moffat, C.F., 2022. The concentration and biomagnification of PCBs and PBDEs across four trophic levels in a marine food web. *Environ. Pollut.* 309, 119752. <https://doi.org/10.1016/j.envpol.2022.119752>.
- Makino, M., 1998. Prediction of n-octanol/waterpartition coefficients of polychlorinated biphenyls by use of computer calculated molecular properties. *Chemosphere* 37 (1), 13–26. [https://doi.org/10.1016/S0045-6535\(98\)00034-4](https://doi.org/10.1016/S0045-6535(98)00034-4).
- Matsumoto, R., Tu, N.P.C., Haruta, S., Kawano, M., Takeuchi, I., 2014. Polychlorinated biphenyl (PCB) concentrations and congener composition in masu salmon from Japan: a study of all 209 PCB congeners by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS). *Mar. Pollut. Bull.* 85 (2), 549–557. <https://doi.org/10.1016/j.marpolbul.2014.04.021>.
- McConnaughey, T., McCoy, R., 1979. Food-web structure and the fractionation of carbon isotopes in the Bering Sea. *Mar. Biol.* 53 (3), 257–262. <https://doi.org/10.1007/BF00952434>.
- Mongillo, T.M., Ylitalo, G.M., Rhodes, L.D., O'Neill, S.M., Noren, D.P., Hanson, M.B., 2016. Exposure to a Mixture of Toxic Chemicals: Implications for the Health of Endangered Southern Resident Killer Whales. <https://doi.org/10.7289/V5/TM-NWFSC-135>.
- Ng, C.A., Ritscher, A., Hungerbuehler, K., von Goetz, N., 2018. Polybrominated diphenyl ether (PBDE) accumulation in farmed salmon evaluated using a dynamic sea-cage production model. *Environ. Sci. Technol.* 52 (12), 6965–6973. <https://doi.org/10.1021/acs.est.8b00146>.
- Niimi, A., Oliver, a.G., 1983. Biological half-lives of polychlorinated biphenyl (PCB) congeners in whole fish and muscle of rainbow trout (*Salmo gairdneri*). *Can. J. Fish. Aquat. Sci.* 40 (9), 1388–1394. <https://doi.org/10.1139/f83-160>.
- O'Neill, S.M., West, J.E., 2009. Marine distribution, life history traits, and the accumulation of polychlorinated biphenyls in chinook salmon from Puget Sound, Washington. *Trans. Am. Fish. Soc.* 138 (3), 616–632. <https://doi.org/10.1577/t08-003.1>.
- O'Neill, S.M., Carey, A.J., Harding, L.B., West, J.E., Ylitalo, G.M., Chamberlin, J.W., 2020. Chemical tracers guide identification of the location and source of persistent organic pollutants in juvenile Chinook salmon (*Oncorhynchus tshawytscha*), migrating seaward through an estuary with multiple contaminant inputs. *Sci. Total Environ.* 712, 135516. <https://doi.org/10.1016/j.scitotenv.2019.135516>.
- O'Neill, S., Ylitalo, G., West, J., Bolton, J., Sloan, C., Krahn, M., 2006. Regional patterns of persistent organic pollutants in five Pacific salmon species (*Oncorhynchus spp*) and their contributions to contaminant levels in northern and southern resident killer whales (*Orcinus orca*). In: 2006 Southern Resident Killer Whale Symposium. NOAA-Fisheries, Northwest Fisheries Science Center, Seattle, WA.
- O'Neill, S.M., Ylitalo, G.M., West, J.E., 2014. Energy content of Pacific salmon as prey of northern and southern resident killer whales. *Endanger. Species Res.* 25 (3), 265–281. <https://doi.org/10.3354/esr00631>.
- Ohlberger, J., Ward, E.J., Schindler, D.E., Lewis, B., 2018. Demographic changes in chinook salmon across the northeast Pacific ocean. *Fish Fish.* 19 (3), 533–546. <https://doi.org/10.1111/faf.12272>.
- Papa, E., Kovarich, S., Gramatica, P., 2009. Development, validation and inspection of the applicability domain of QSPR models for physicochemical properties of polybrominated diphenyl ethers. *QSAR Comb. Sci.* 28 (8), 790–796. <https://doi.org/10.1002/qsar.200860183>.
- Pearce, R., 2018. Bioaccumulation of PCBs in Southern Resident Killer Whales in the Salish Sea.
- Post, D.M., Layman, C.A., Arrington, D.A., Takimoto, G., Quattrochi, J., Montana, C.G., 2007. Getting to the fat of the matter: models, methods and assumptions for dealing with lipids in stable isotope analyses. *Oecologia* 152, 179–189. <https://doi.org/10.1007/s00442-006-0630-x>.
- Quinn, T.P., 2018. The abundance and diversity of salmon and trout: Past, present, future. In: *The Behavior and Ecology of Pacific Salmon and Trout*. University of Washington Press.
- R Core Team, 2020. R: a language and environment for statistical computing. In: R Foundation for Statistical Computing. <https://www.R-project.org/>.
- Riddell, B., Bradford, M., Carmichael, R., Hankin, D., Peterman, R., Wertheimer, A., 2013. Assessment of Status and Factors for Decline of Southern BC Chinook Salmon: Independent Panel's Report. Prepared with the assistance of Marmorek DR, & Hall, AW ESSA Technologies Ltd., Vancouver, BC for Fisheries and Oceans Canada (Vancouver, BC) and Fraser River Aboriginal Fisheries Secretariat (Merritt, BC).
- Roberts, S.C., Noyes, P.D., Gallagher, E.P., Stapleton, H.M., 2011. Species-specific differences and structure–activity relationships in the debromination of PBDE congeners in three fish species. *Environ. Sci. Technol.* 45 (5), 1999–2005. <https://doi.org/10.1021/es103934x>.
- Ross, P.S., 2006. Fireproof killer whales (*Orcinus orca*): flame-retardant chemicals and the conservation imperative in the charismatic icon of British Columbia, Canada. *Can. J. Fish. Aquat. Sci.* 63 (1), 224–234. <https://doi.org/10.1139/f05-244>.
- Ross, P.S., Ellis, G., Ikonomou, M., Barrett-Lennard, L., Addison, R., 2000. High PCB concentrations in free-ranging Pacific killer whales, *Orcinus orca*: effects of age, sex and dietary preference. *Mar. Pollut. Bull.* 40 (6), 504–515. [https://doi.org/10.1016/S0025-326X\(99\)00233-7](https://doi.org/10.1016/S0025-326X(99)00233-7).
- Ross, P.S., Jeffries, S.J., Yunker, M.B., Addison, R.F., Ikonomou, M.G., Calambokidis, J. C., 2004. Harbor seals (*Phoca vitulina*) in British Columbia, Canada, and Washington State, USA, reveal a combination of local and global polychlorinated biphenyl, dioxin, and furan signals. *Environ. Toxicol. Chem.: Int. J.* 23 (1), 157–165. <https://doi.org/10.1897/03-85>.
- Ross, P.S., Noël, M., Lambourn, D., Dangerfield, N., Calambokidis, J., Jeffries, S., 2013. Declining concentrations of persistent PCBs, PBDEs, PCDEs, and PCNs in harbor seals (*Phoca vitulina*) from the Salish Sea. *Prog. Oceanogr.* 115, 160–170. <https://doi.org/10.1016/j.pocean.2013.05.027>.
- Secretariat of the Stockholm Convention, 2019. The POPs. United Nations Environment Programme Stockholm Convention. Retrieved January 17 from. <http://chm.pops.int/theconvention/the pops/tabid/673/default.aspx>.
- Sharma, R., Quinn, T.P., 2012. Linkages between life history type and migration pathways in freshwater and marine environments for Chinook salmon, *Oncorhynchus tshawytscha*. *Acta Oecol.* 41, 1–13. <https://doi.org/10.1016/j.actao.2012.03.002>.
- Shaw, S.D., Berger, M.L., Brenner, D., Carpenter, D.O., Tao, L., Hong, C.S., Kannan, K., 2008. Polybrominated diphenyl ethers (PBDEs) in farmed and wild salmon marketed in the Northeastern United States. *Chemosphere* 71 (8), 1422–1431. <https://doi.org/10.1016/j.chemosphere.2008.01.030>.
- Shedd, K.R., Leonard, D.L., Nichols, J.V., 2021. Mixed Stock Analysis of Chinook Salmon Harvested in Southeast Alaska Commercial and Troll Sport Fisheries, 2018. Alaska Department of Fish and Game, Fishery Data Series No. 21-23, Anchorage.
- Shedd, K.R., Leonard, D.L., Nichols, J.V., 2022. Mixed Stock Analysis of Chinook Salmon Harvested in Southeast Alaska Commercial Troll and Sport Fisheries, 2019. Alaska Department of Fish and Game, Fishery Data Series No. 22-20, Anchorage.
- Shelton, A.O., Satterthwaite, W.H., Ward, E.J., Feist, B.E., Burke, B., 2019. Using hierarchical models to estimate stock-specific and seasonal variation in ocean distribution, survivorship, and aggregate abundance of fall run Chinook salmon. *Can. J. Fish. Aquat. Sci.* 76 (1), 95–108. <https://doi.org/10.1139/cjfas-2017-0204>.
- Simenstad, C.A., Fresh, K.L., Salo, E.O., 1982. The role of puget sound and Washington coastal estuaries in the life history of pacific salmon: an unappreciated function. In: Kennedy, V.S. (Ed.), *Estuarine Comparisons*. Academic Press, pp. 343–364. <https://doi.org/10.1016/B978-0-12-404070-0.50026-0>.
- Stone, D., 2006. Polybrominated diphenyl ethers and polychlorinated biphenyls in different tissue types from Chinook salmon (*Oncorhynchus tshawytscha*). *Bull. Environ. Contam. Toxicol.* 76 (1), 148–154. <https://doi.org/10.1007/s00128-005-0901-y>.
- Tucker, S., Trudel, M., Welch, D.W., Candy, J.R., Morris, J.F.T., Thiess, M.E., Wallace, C., Beacham, T.D., 2012. Annual coastal migration of juvenile Chinook salmon: static stock-specific patterns in a highly dynamic ocean. *Mar. Ecol. Prog. Ser.* 449, 245–262. <https://doi.org/10.3354/meps09528>.
- Veldhoen, N., Ikonomou, M.G., Dubetz, C., Macpherson, N., Sampson, T., Kelly, B.C., Helbing, C.C., 2010. Gene expression profiling and environmental contaminant assessment of migrating Pacific salmon in the Fraser River watershed of British Columbia. *Aquat. Toxicol.* 97 (3), 212–225. <https://doi.org/10.1016/j.aquatox.2009.09.009>.
- Waples, R.S., 1991. Pacific salmon, *Oncorhynchus spp.*, and the definition of "species" under the Endangered Species Act. *US Natl. Mar. Fish. Serv. Mar. Fish. Rev.* 53 (3), 11–22.
- Ward, E., Hanson, M., Weitkamp, L., Ford, M., Fisheries, N., 2010. Modeling Killer Whale Prey Size Selection Based upon Available Data. Northwest Fisheries Science Center.
- Weitkamp, L.A., 2010. Marine distributions of chinook salmon from the West Coast of North America determined by coded Wire Tag Recoveries. *Trans. Am. Fish. Soc.* 139 (1), 147–170. <https://doi.org/10.1577/t08-225.1>.
- West, J.E., O'Neill, S.M., Ylitalo, G.M., 2008. Spatial extent, magnitude, and patterns of persistent organochlorine pollutants in Pacific herring (*Clupea pallasii*) populations in the Puget Sound (USA) and Strait of Georgia (Canada). *Sci. Total Environ.* 394 (2–3), 369–378. <https://doi.org/10.1016/j.scitotenv.2007.12.027>.
- West, J.E., O'Neill, S.M., Ylitalo, G.M., 2017. Time trends of persistent organic pollutants in benthic and pelagic indicator fishes from Puget Sound, Washington, USA. *Arch. Environ. Contam. Toxicol.* 73 (2), 207–229. <https://doi.org/10.1007/s00244-017-0383-z>.
- Williams, T.H., Spence, B.C., Boughton, D.A., Johnson, R.C., Crozier, E.G.R., Mantua, N. J., O'Farrell, M.R., Lindley, S.T., 2016. Viability Assessment for Pacific Salmon and Steelhead Listed under the Endangered Species Act: Southwest.
- World Health Organization, 1994. Brominated Diphenyl Ethers, 9241571624.
- Xu, Y., Scott Decker, A., Parken, C.K., Ritchie, L.M., Patterson, D.A., Fu, C., 2020. Climate effects on size-at-age and growth rate of Chinook Salmon (*Oncorhynchus tshawytscha*) in the Fraser River, Canada. *Fish. Oceanogr.* 29 (5), 381–395. <https://doi.org/10.1111/fo.12484>.
- Yunker, M.B., Ikonomou, M.G., Sather, P.J., Friesen, E.N., Higgs, D.A., Dubetz, C., 2011. Development and validation of protocols to differentiate PCB patterns between farmed and wild salmon. *Environ. Sci. Technol.* 45 (6), 2107–2115. <https://doi.org/10.1021/es1038529>.
- Zanden, M.J.V., Rasmussen, J.B., 2001. Variation in $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ trophic fractionation: implications for aquatic food web studies. *Limnol. Oceanogr.* 46 (8), 2061–2066. <https://doi.org/10.4319/lo.2001.46.8.2061>.