

Marine Distribution, Life History Traits, and the Accumulation of Polychlorinated Biphenyls in Chinook Salmon from Puget Sound, Washington

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Abstract.—Polychlorinated biphenyl (PCB) levels and the factors affecting PCB accumulation in subadult and maturing Chinook salmon *Oncorhynchus tshawytscha* from Puget Sound were characterized. Specifically, we (1) determined PCB levels in Chinook salmon from Puget Sound and compared them with levels in Chinook salmon from other West Coast populations, (2) determined whether PCB accumulation mainly occurred in the freshwater or marine habitats, and (3) quantified the relative importance of fish age, fish size (fork length), lipid content, and saltwater age (the number of winters spent in saltwater) on PCB concentration. The average PCB concentration measured in skinless muscle tissue samples of subadult and maturing Chinook salmon collected from Puget Sound was 53 ng/g (wet weight), which was 3–5 times higher than those measured in six other populations of Chinook salmon on the West Coast of North America. Concentrations in the Puget Sound samples varied from 10 to 220 ng/g. A comparison of PCB body burdens between subyearling smolts and returning adults revealed that almost all of the PCBs (>96%) were accumulated in the marine habitats. Surprisingly, although PCBs were mostly accumulated in marine habitats, PCB exposure was lowest in the largest fish that spent the most time in saltwater. Collectively, saltwater age, fish size, and lipids only accounted for 37% of the observed variation in PCB concentration, indicating that some other attribute of the fish's marine ecology accounted for the variation in PCB levels among Puget Sound Chinook salmon and for their elevated PCB levels relative to other West Coast populations. We hypothesized that residency in the contaminated Puget Sound environment was a major factor contributing to the higher and more variable PCB concentrations in these fish. This hypothesis was supported with an independent data set from a fishery assessment model, which estimated that 29% of subyearling Chinook salmon and 45% of yearling out-migrants from Puget Sound displayed resident behavior.

Polychlorinated biphenyls (PCBs) are among the most ubiquitous and persistent contaminants in aquatic ecosystems worldwide (Phillips 1994). Although their production in North America has been banned since the mid 1970s, their persistence and toxicity continues to be a cause for concern in aquatic species, especially fishes, and the wildlife and humans that eat them. In populated areas, PCBs entered aquatic ecosystems via spills, surface runoff, and groundwater leaching and then accumulated in the biota. Polychlorinated biphenyls are transported to remote, otherwise uncontaminated areas via atmospheric processes (Hammar 1989; Wania and Mackay 2001), but they can also be transported in the bodies of animals migrating between more- and less-contaminated areas (Ewald et al. 1998; Krümmel et al. 2003; Blais 2005).

The accumulation of PCBs in fishes depends

primarily on their proximity to contaminated habitats and the PCB levels in their food (Thomann and Connolly 1984; Russell et al. 1999), but other traits such as life span (or duration of exposure), trophic status, growth rate, lipid content, gender, and age of first reproduction can also exacerbate or mitigate PCB accumulation (Jensen et al. 1982; Rasmussen et al. 1990; Larsson et al. 1991, 1993, 1996; Hammar et al. 1993; Madenjian et al. 1994, 1998; Stow 1995; Bentzen et al. 1996; Johnston et al. 2002). Thus, studies evaluating the relationship between proximity to contaminated habitats and PCB accumulation by fish in these habitats must fully consider how these traits may affect the observed PCB levels. For migratory fishes like Pacific salmon *Oncorhynchus* spp., the link between contaminated habitats and PCB accumulation in fish tissue can be further obscured by the complex movement patterns within and between freshwater and marine habitats, which may differ dramatically in contaminant levels.

Polychlorinated biphenyls have been detected in Pacific salmon from remote and populated locations in the northeast Pacific Ocean (Stout and Beezhold 1981; Tetra Tech, Inc. et al. 1996; Ewald et al. 1998; O'Neill

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et al. 1998; USEPA 2002; West et al. 2002; Hites et al. 2004; Missildine et al. 2005; Rice and Moles 2006; Ikonomou et al. 2007), but the complex life history and long-range feeding migrations of salmon have hindered a full understanding of the factors affecting their PCB accumulation. Pacific salmon spawn in freshwater, where the juveniles live for a few days to two or more years depending on species and population. The salmon then migrate to marine waters, where they achieve more than 99% of their final adult mass; they subsequently return to their natal freshwater habitat to spawn (Quinn 2005). The marine distribution and duration of residence in saltwater vary widely between species and between populations within species (Quinn and Myers 2004; Quinn 2005). Thus, Pacific salmon may be exposed to PCBs in freshwater, estuarine, and marine habitats, and throughout their geographic range the levels of PCB contamination in salmon habitats may vary widely. Lipophilic compounds like PCBs readily accumulate in muscle tissue of adult Pacific salmon because of their relatively high fat content. However, species and populations differ in their fat content depending on their diet in saltwater and the migratory pattern they undertake (Brett 1995; Quinn 2005). Furthermore, the lipid content in the muscle tissue of adult salmon in marine waters decreases rapidly as they approach freshwater and reproductive maturity (Brett 1995; Ewald et al. 1998; Hendry and Berg 1999). During this reproductive phase, PCBs are not metabolized with the fat but instead are mobilized and redistributed to fatter tissues, such as the gonads (Ewald et al. 1998; Kelly et al. 2007). Therefore, inter- and intraspecific differences in duration of exposure in the freshwater, estuarine, and marine habitats and differences in the diet and lipid content of the fish can all influence PCB concentrations in Pacific salmon muscle tissue.

Like many estuaries along the West Coast of North America, the inland marine and estuarine ecosystems of Washington and British Columbia have been altered dramatically over the last 100 years by anthropogenic activities, including overfishing, habitat loss, and inputs of toxic chemicals (Wilson et al. 1994; West 1997). This region (Figure 1) comprises an extensive series of relatively deep, fjord-like basins, including the Strait of Georgia, the Strait of Juan de Fuca, and Puget Sound. In both Puget Sound and the Strait of Georgia, exposure of biota to anthropogenic toxic contaminants is of particular concern because the region is experiencing rapid human population growth, and the enclosed nature of its inland marine and estuarine waters may impede the dilution of contaminants (Harrison et al. 1994). In particular, fish and wildlife that feed in Puget Sound are at greater risk of exposure

to contaminants than those feeding in the Strait of Georgia because the Puget Sound watershed is more densely populated and its shoreline is more developed with industry and urban centers. Puget Sound is also a much smaller body of water; its surface area is only one-third that of the Strait of Georgia, its volume is only one-sixth that of the strait, and the summer residence time of water is roughly double that of the strait (adapted from Thomson 1994). Indeed, West et al. (2008) documented that the Pacific herring *Clupea pallasii* populations in Puget Sound were 3–9 times more contaminated with PCBs than those from the Strait of Georgia.

Chinook salmon *O. tshawytscha* originating in rivers of Puget Sound are of special concern with respect to contaminants for several reasons. First, this species is extensively targeted in commercial and recreational fisheries; thus, the safety of food for human consumption is an issue. Second, the Puget Sound Chinook salmon Evolutionarily Significant Unit is listed as threatened under the U.S. Endangered Species Act (ESA), so factors such as contaminants that might contribute to their decline or hinder their recovery are important. Finally, Chinook salmon are key prey of southern resident killer whales *Orcinus orca* (Ford 1998; Ford and Ellis 2006), and contaminant transfer to killer whales via their predation on salmon may have contributed to the decline of this whale population to the point where they are now also listed under the ESA (Krahn et al. 2004; U.S. Department of Commerce 2005).

Polychlorinated biphenyls have been documented in skinless fillets taken from adult Chinook salmon returning to streams in the Puget Sound region (O'Neill et al. 1998; West et al. 2002; Missildine et al. 2005), but salmon life history traits (e.g., age and size at maturity) and migration patterns that might affect PCB concentration have not been adequately evaluated. In particular, it has long been known that some Chinook salmon spend all or most of their marine life in the inland marine waters of Washington and British Columbia instead of migrating to feed on the continental shelf as is more typical of the species. These fish, termed "residents," are caught within the inland marine waters during times of the year (e.g., from late fall to early spring) when most conspecifics are feeding in coastal or oceanic regions of the Pacific Ocean (Pressey 1953; Haw et al. 1967). Residents could thus be exposed to higher levels of PCBs and other contaminants because their marine distribution is more restricted than that of migratory Chinook salmon.

The objective of this study was to characterize the effects of life history traits on PCB levels in Puget Sound Chinook salmon. Specifically, we first tested the null hypothesis that PCB concentrations measured in

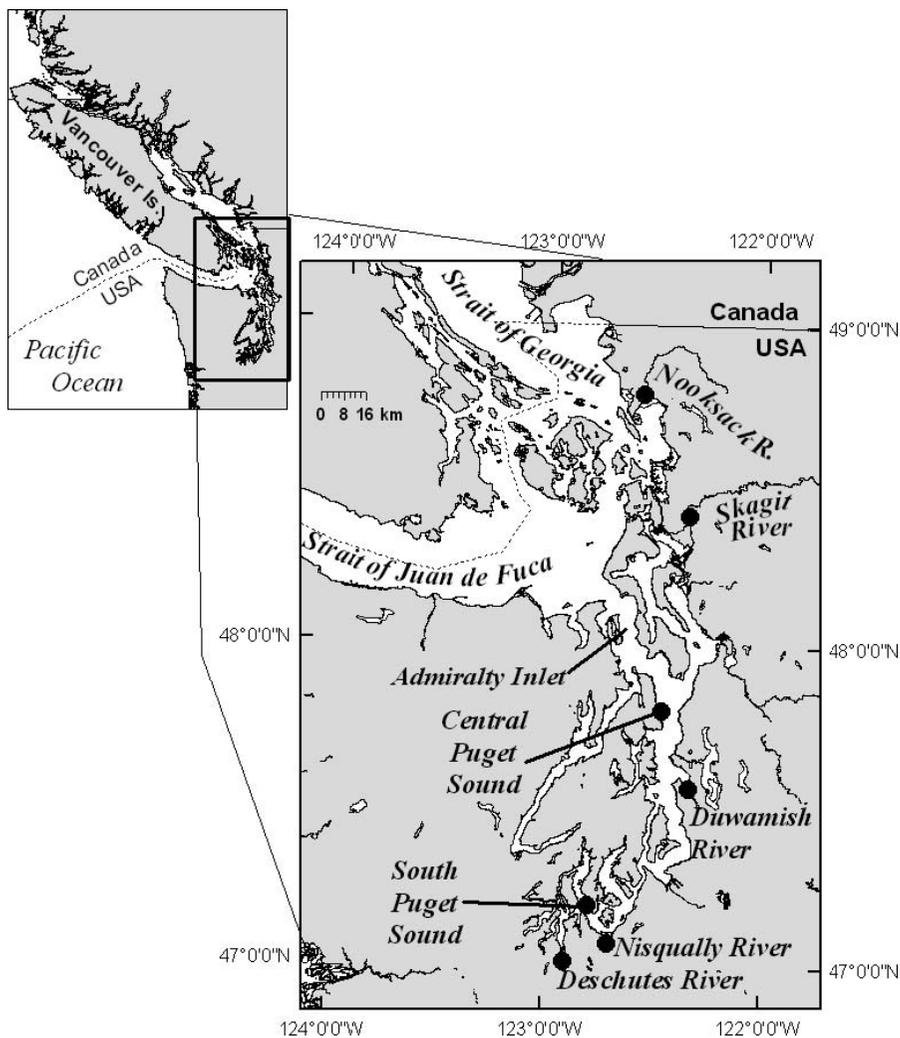


FIGURE 1.—Sampling locations for Chinook salmon in Puget Sound, Washington, 1992–1996. Inset shows location of Puget Sound and adjacent waters on the West Coast of USA–Canada.

Chinook salmon originating from Puget Sound would be similar to published values for other North American West Coast populations; this hypothesis was evaluated against the predicted alternative, which is that values for Puget Sound Chinook salmon would be higher than those for other West Coast populations. Second, we tested whether PCB accumulation occurred mainly in the freshwater or marine habitats by comparing the PCB body burdens (total ng/fish) in juveniles migrating from a highly contaminated river with the body burdens in adults returning to that same river. Third, we tested the hypotheses that fish size, age, and lipid content affect PCB concentration, and we determined the relative importance of these factors.

Fourth, we used a fishery assessment model based on recoveries of tagged fish during routine fishery sampling to estimate the proportion of Chinook salmon that reside in Puget Sound instead of migrating to the Pacific Ocean. We discuss the role that residency in Puget Sound may play in PCB concentration for Chinook salmon populations from Puget Sound compared with other populations originating from less-contaminated regions and the implications of PCB contamination for the health of Puget Sound Chinook salmon and their predators.

Methods

Sampling locations.—In August and September 1992–1996, maturing and subadult Chinook salmon

were sampled from the inland marine and estuarine waters of Washington (Figure 1), hereafter referred to as Puget Sound, which includes the marine waters south of Admiralty Inlet, U.S. boundary waters of the Strait of Juan de Fuca, and southern portions of the Strait of Georgia (Puget Sound Action Team 2002). Six-hundred thirty-four Chinook salmon were sampled from five "in-river" locations (Nooksack, Skagit, Duwamish, Nisqually, and Deschutes rivers), including nearshore estuarine and riverine areas, where sexually mature captured fish were presumed to be returning to their natal streams to spawn and thus represented distinct populations. In addition, 129 Chinook salmon were sampled from two marine locations, one in central Puget Sound and one in southern Puget Sound, where the natal stream of each fish was unknown (Figure 1). These samples included (1) maturing fish that would have spawned in their year of capture and (2) immature subadult fish that would have spawned in subsequent years.

Each in-river location was sampled in at least 3 years, and each marine location was sampled at least twice. Most fish were purchased from licensed commercial fish buyers and treaty tribal fishermen, but Chinook salmon returning to the Deschutes River were collected by Washington Department of Fish and Wildlife (WDFW) staff at a trap designed to collect returning mature fish. Fish sampled from central Puget Sound were collected from a WDFW test fishery.

Sample preparation.—Whole salmon were transported on ice to the laboratory, where they were measured for fork length (FL, mm) and mass (g), and scales were removed for age estimation. The fish were then wrapped individually in aluminum foil, placed in plastic bags, and stored on ice for up to 10 d until tissues were removed for contaminant analyses. The total age of each fish and the year in which it migrated to saltwater were estimated based on the pattern and spacing of circuli on the scales. This standard methodology is typically more than 95% accurate for adult salmon (e.g., 97% for Chinook salmon based on recoveries of tagged fish; Unwin and Lucas 1993; M. Unwin, National Institute of Water and Atmospheric Research, Christchurch, New Zealand, personal communication). Most (70%) of the Chinook salmon migrated to saltwater as subyearlings (i.e., spent less than one full year in freshwater), and 30% were yearlings (i.e., spent one full year plus a few months in freshwater). The saltwater age (SWA) of the fish, defined as the number of full winters spent in saltwater, was calculated for each fish by subtracting the number of partial years the fish spent in freshwater from their total age. The pattern of circuli in the freshwater zone of the scale could only be used to classify

hatchery or wild origin (Unwin and Lucas 1993) for the fish that migrated to saltwater as yearlings (30% of total sampled fish), and 98% of these were hatchery fish. Thus, a comparison of PCB levels between hatchery and wild fish was not possible.

In total, 204 muscle tissue samples were obtained from the 763 individual Chinook salmon collected from Puget Sound. Individual muscle tissue samples were collected from 50 fish; the remaining 713 fish were combined to create 154 composite samples of muscle tissue. Composite samples were prepared by collecting equal amounts of skinned muscle tissue from two to five individuals. In 1992 and 1993, fish from one location were combined randomly into composite samples without consideration of fish age or out-migrant life history to create 60 composite samples (i.e., random fish age composites). To better assess the relationship between fish life history and PCB concentration, most (74 of 94) muscle tissue samples at each location from 1994 to 1996 were composites of fish with the same age and life history (i.e., uniform fish age composites) or were obtained from individual fish (50 samples). Random fish age composites (1992–1993) and uniform fish age composites (1994–1996) were collected at all seven sampling locations (five in-river locations and two marine locations). Individual fish samples (1994–1996) were collected at four of the five in-river locations. For composite samples, the total age, SWA, and FL of fish in the sample were represented by the mean total age, mean SWA, and mean FL of fish in that sample.

In general, compositing should dampen the variability associated with PCB concentration among individuals and is a suitable approach to represent the central tendency of the Puget Sound population. The variability among random fish age composites should be less than that observed for the uniform fish age composites, and the highest variability should be observed for the individual samples. However, given that the main objective of this paper was to characterize factors affecting PCB accumulation in Puget Sound Chinook salmon as a whole rather than to examine the differences among populations, the compositing procedure as executed should not affect the overall results.

Chemical analyses.—All muscle tissue samples were analyzed for the presence of lipids and PCBs, measured as Aroclor 1016, 1221, 1232, 1242, 1248, 1254, and 1260, at the King County Environmental Laboratory in Seattle, Washington, using standard U.S. Environmental Protection Agency methods 8080 and 8082. Aroclor analyses were conducted instead of PCB homologue or PCB congener analyses because at the time this study was completed, the Aroclor method was the most

common method for measuring total PCBs due to cost, equipment availability, time, and technology.

Tissue samples delivered to the chemical laboratory were homogenized in a blender and then stored at -20°C until analyzed. Homogenized, 30-g tissue samples were mixed with anhydrous sodium sulfate, spiked with surrogates, and extracted with a sonic probe using a 1:1 mixture of methylene chloride and acetone. Portions of the extracts were analyzed separately for PCBs and lipids.

Polychlorinated biphenyl extracts were "cleaned" by gel permeation chromatography and concentrated to a final volume of 1 mL. The PCBs were analyzed using gas chromatography–electron capture detection with either a dual narrow-bore column (0.25 mm) suited to analyzing low concentrations (1992–1994) or an ion trap detector (1995 and 1996). The identification of Aroclors in the tissue samples was accomplished by pattern recognition, and retention times were compared with standards for PCB Aroclor mixtures. Five-point calibration curves were prepared for Aroclors 1016 and 1260. Three-point calibration curves were prepared for Aroclors 1221, 1232, 1242, 1248, and 1254. Calibration checks were analyzed daily with every batch of 20 or fewer samples. A group of peaks in both the samples and the standards were summed to quantitate the PCBs, and an external standard was used to calculate the final volume. Matrix-based detection limits (~ 2 ng/g for individual Aroclors) were determined for the muscle tissue by adding standards to representative instrument-ready sample matrices. Total PCBs for muscle tissue samples were calculated by summing the detected values for Aroclors 1254 and 1260, which were detected in 100% and 99% of the samples, respectively. No other Aroclors were detected. All PCB values were reported as the concentration per wet weight of tissue in nanograms per gram (ng/g).

A method blank, a spiked method blank, a matrix spike, a matrix spike duplicate, and laboratory duplicate were analyzed for each batch of 20 or fewer samples, and all were within the standard limits of acceptability. We measured the recovery of surrogate compounds (2,4,5,6 tetrachlor-*m*-xylene and decachlorobiphenyl) that had been added to all muscle tissue samples and quality control samples to monitor performance. Additionally, as a general indication of laboratory accuracy, at least one Standard Reference Material (SRM 1588 [cod liver oil], SRM Muscle Tissue V, or both) was analyzed for the presence of three PCB isomers for every 50 samples analyzed, and all were within the standard limits of acceptability.

The percent lipid of each muscle tissue sample was determined by drying the extract over anhydrous sodium sulfate and then evaporating the samples to

dryness in a tared weighing vessel. The extract was prepared as described previously and was filtered on a bed of sodium sulfate powder; the solvent was evaporated to dryness in a tared weighing vessel. The resulting residues were dried to a constant weight, and the lipid percentage was then determined gravimetrically and reported as percent of total weight. For composite samples, this represents the mean percent lipids for all fish in the composite because an equal mass of tissue was taken from each fish.

Comparison of PCB accumulation by Chinook salmon in freshwater and marine habitats.—Total PCB body burdens (ng/fish) in out-migrating smolts and returning adults were compared to assess the percent of the total PCB accumulation acquired while rearing in the freshwater habitat. Chinook salmon from the highly industrialized Duwamish River were selected to estimate the maximum contribution from freshwater because this river has some of the most PCB-contaminated juvenile salmonids in Puget Sound and the Pacific Northwest (Varanasi et al. 1993; Meador et al. 2002; Johnson et al. 2007). Body burdens were calculated as:

$$\text{PCB body burden (ng/fish)} \\ = [\text{whole-body PCB (ng/g)}] \times [\text{fish weight (g)}].$$

Whole-body PCB concentrations and weights for out-migrating smolts were provided by J. P. Meador (National Oceanic and Atmospheric Administration [NOAA] Fisheries, Northwest Fisheries Science Center, Seattle, Washington, personal communication). These PCB data were consistent with other published PCB concentrations for Duwamish River fish (Varanasi et al. 1993; Meador et al. 2002; Johnson et al. 2007); however, this data set was selected for the analysis because the associated smolt weights were available, whereas weights were not reported in the published studies. Whole-body PCB concentrations for returning adults were estimated from muscle tissue concentrations using the muscle–fillet relationship generated from 10 paired muscle and whole-body samples with fillet concentrations ranging from 12 to 210 ng/g ($r^2 = 0.96$, $P < 0.0001$):

$$\text{Adult whole-body PCB (ng/g)} \\ = \{1.074 \times [\text{fillet PCB (ng/g)}]\} + 0.8993.$$

These 10 paired samples were collected in 2000 and were considered to be generally representative of the relationship between muscle PCB and whole-body PCB concentrations for Puget Sound Chinook salmon because they were collected during the same time of year (August–September) as in the current study and included two of the same Puget Sound populations

(Nooksack and Nisqually rivers). Fish in the paired samples ranged from 2 to 4 years of age, and the muscle lipid content average (5.4%) and range (2.7–8.0%) were similar to those observed for the 1992–1996 sampling.

Whole-body PCB concentrations for out-migrating smolts were only available for subyearling smolts, so the comparison of PCB body burden between smolts and adults was also limited to adults that migrated to saltwater as subyearlings. For each adult returning to the Duwamish River, the maximum contribution from freshwater was calculated as the 95th-percentile PCB body burden (ng/fish) of smolts divided by the PCB body burden of adults.

Estimates of residency by Puget Sound Chinook Salmon.—The degree to which Puget Sound Chinook salmon reside in Puget Sound, where exposure to PCBs may be greater than that for fish migrating to the Pacific Ocean, is unknown. We used population-specific catch estimates generated from the Fishery Regulation Assessment Model (FRAM; Pacific Fisheries Management Council 2006) to infer the percentage of fish of Puget Sound origin that resided in the inland marine waters of Washington and British Columbia in general (i.e., WDFW management areas 5–13 and Canadian waters in the Strait of Juan de Fuca and Strait of Georgia) and in inner Puget Sound specifically (i.e., the area south of Admiralty Inlet; WDFW management areas 9–11 and 13). This fishery simulation model is based on the dates and locations of recoveries of known-origin salmon (tagged with coded wire tags before release from hatcheries) in fisheries from Alaska to California and is used by the WDFW and other management agencies to assess fishery impacts on hatchery and wild populations of Chinook salmon and coho salmon *O. kisutch* along the West Coast of North America. We used the FRAM to estimate the numbers of Puget Sound-origin Chinook salmon that were landed in area-specific fisheries during 1992 through 1996, when we sampled fish for our study. Within each year, estimates of landings were stratified by catch area, age-class, and season (i.e., October–April, May–June, and July–September). Catch areas constituting the inland marine waters of British Columbia and Washington included the Strait of Juan de Fuca, Strait of Georgia, Hood Canal, and inner Puget Sound. Catch areas outside the inland marine waters of Washington and British Columbia included waters of Alaska, northern British Columbia, the west coast of Vancouver Island, and coastal waters of Washington, Oregon, and California.

Chinook salmon that originated in Puget Sound watersheds were classified as having “resident” behavior if they were caught in the inland waters

during October through June, outside the typical July–September migration timing for the most abundant summer–fall migrant adults returning from the ocean to Puget Sound to spawn. We did not account for the small numbers of spring migrant populations of Chinook salmon that might have been present in Puget Sound because they constitute less than 5% of the total population. Although fish caught in the winter fisheries were classified as residents at their time of capture, the proportion of their lives spent in the inland waters, particularly Puget Sound, is unknown. The recovery of fish in a fishery is subject to variations in annual and seasonal regulations; hence, the results are subject to all of the biases associated with fisheries, such as time, area, and gear restrictions. However, the inferences drawn from this model were sufficiently limited that it was robust for classifying resident behavior.

Statistical analysis.—A one-way analysis of variance (ANOVA; Sigma Plot 2006) was used to test for significant differences in PCB concentration and sample fish FL among locations, with log transformation used to normalize data as necessary. Tukey’s post hoc multiple range tests were run for all significant differences to determine which locations differed from each other.

A stepwise general linear model (GLM; Systat 2000) was used to evaluate whether SWA contributed significantly to the variability in observed PCB concentrations, while accounting for potential effects related to sample fish FL and percent lipids. The goal was to derive a predictive regression model between PCB concentration and SWA that reduced any statistically significant effects of FL and percent lipids covariates. We computed multiple GLMs beginning with the most complex model that included SWA, both covariates, and all interactions; we then iteratively removed interaction terms or covariates with a *P*-value less than 0.05 in a stepwise fashion starting with the three-way interaction. In this study, no interaction term was significant regardless of the order of stepwise removal. The final model calculated a predicted PCB concentration for each SWA group based on the grand mean of the significant covariates and tested the significance of differences in these predicted concentrations.

Additionally, a GLM was also used to test whether PCB concentration varied among sampling locations while holding SWA constant (selecting only fish that had spent 2 years in saltwater; i.e., SWA2) and adjusting for fish size and lipid differences using the same stepwise procedure as above. We also used a GLM to test whether PCB concentration in subadults and adults was affected by the time they spent in

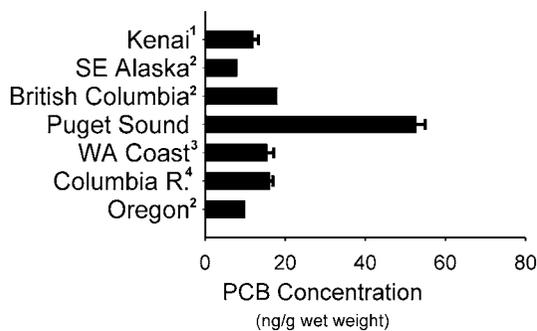


FIGURE 2.—Average (\pm SE) PCB concentration in Chinook salmon filets. Data for Puget Sound were based on 204 samples collected by the Washington Department of Fish and Wildlife from 1992 to 1996; data for other locations were taken from the following (indicated by superscript numbers): ¹Rice and Moles (2006), ²Hites et al. (2004; estimated from publication), ³Missildine et al. (2005), and ⁴U.S. Environmental Protection Agency (USEPA 2002).

freshwater (i.e., subyearlings or yearlings) while holding SWA constant and adjusting for fish size.

Results and Discussion

PCB Concentrations in Chinook Salmon from Puget Sound and Other West Coast Populations

The average concentration of PCBs measured in 204 samples of skinless muscle tissue from subadult and maturing Chinook salmon from Puget Sound was 53 ng/g fish tissue, which was three to five times higher than average concentrations reported for adult Chinook salmon from six other populations on the West Coast of North America (Figure 2). The PCBs in Puget Sound Chinook salmon varied widely among samples, from 10 to 220 ng/g, and significant differences were evident among the seven sampling locations (ANOVA on \log_e [PCBs]: $P < 0.001$). The highest average concentrations were observed in fish caught at the two marine locations (central and south Puget Sound), followed by in-river samples from the Deschutes, Duwamish, and Nisqually rivers (Figure 3, Table 1). Fish from the Nooksack and Skagit rivers had the lowest PCB concentrations (Figure 3). Despite the variation in fish PCB concentrations among Puget Sound sampling locations, all Puget Sound samples had higher average concentrations than samples obtained outside of Puget Sound.

The higher observed PCB levels in Puget Sound fish compared with other West Coast populations were probably not due to differences in biological traits, such as lipid content, fish age, and fish size. Previous studies have documented a strong positive relationship between average lipid content and average total PCB concentration across species in Lake Michigan salmo-

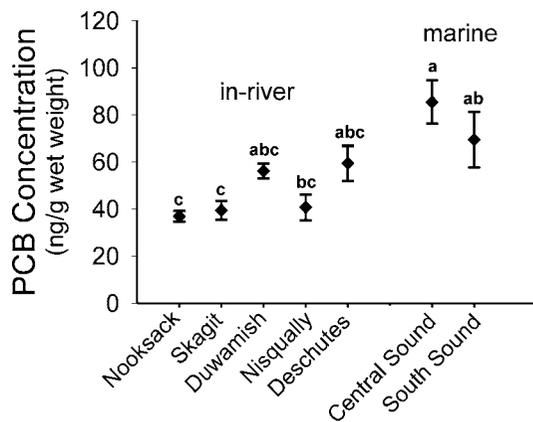


FIGURE 3.—Arithmetic mean (\pm SE) PCB concentration in adult Chinook salmon returning to Puget Sound rivers (in-river) and in those collected from marine areas (marine) in central and southern Puget Sound, Washington. Lowercase letters denote statistically significant groupings based on Tukey's post hoc multiple range test; points sharing letters in common are not significantly different.

nids but only a weak positive relationship within species (Stow et al. 1997; Amrhein et al. 1999). Lipid levels were not reported in most of the PCB studies used for comparison with our samples. However, the lipid content we measured for maturing Puget Sound Chinook salmon was generally lower than those reported for other West Coast populations (Carlson and Hites 2005; Cullon et al. 2009). Thus, if other factors like diet and environmental contamination were the same across populations, the lower lipid content of Puget Sound fish would tend to slightly decrease their PCB levels and thus cannot be responsible for the observed patterns.

Assuming a consistent contaminant source, fish generally bioaccumulate PCBs as they age such that the older and larger fish within a species will accumulate higher concentrations. Although information on the size or age of the fish was not provided for most PCB studies we used for comparison with our results, Puget Sound Chinook salmon generally do not live as long as most populations from Alaska and British Columbia (Myers et al. 1998). Therefore, one would expect lower PCB concentrations for Puget Sound fish, but we observed the opposite (Figure 2). Furthermore, Missildine et al. (2005) documented that PCB concentrations in hatchery Chinook salmon from Puget Sound were almost three times higher than those in coastal Washington hatchery fish of the same age, even though the coastal populations had higher lipid contents; those authors concluded that the difference was due to a higher PCB source in Puget Sound or

TABLE 1.—Number of Chinook salmon collected (*Nf*) and aged (*Nfa*) and the percentage (%) of aged fish that spent one winter in saltwater (% SWA1) from seven sampling locations in Puget Sound, Washington; and the average PCB concentration (wet weight), sample percent lipids (PL), sample fish age (FA), sample fish saltwater age (SWA), and sample fish length (FL) of Chinook salmon sampled for muscle tissue from each sampling location. Fish were collected in August and September from 1992 to 1996 and included both subyearling and yearling out-migrants. At each sampling location, the collected fish were combined to create the number of muscle samples (*Ns*) for chemical analyses and included samples of individual fish and composite samples of 2–5 individuals.

Location type	Puget Sound basin	Location	<i>Nf</i>	<i>Nfa</i>	% SWA1	<i>Ns</i>	PCBs (ng/g)	PL (%)	FA (years)	SWA (years)	FL (mm)
In-river	North	Nooksack River	133	120	3.3	28	37	3.45	3.6	2.5	741
		Skagit River	125	114	3.5	29	40	4.83	4.1	2.6	816
	Central	Duwamish River	171	159	12.6	65	56	7.34	3.8	2.4	763
	South	Nisqually River	92	90	5.6	20	41	3.76	3.4	2.3	732
	South	Deschutes River	113	77	0.0	34	59	1.74	3.9	2.4	789
		All in-river sites	634	560	5.9	176	49	4.82	3.8	2.4	770
Marine	Central	Central sound	60	60	76.7	12	86	5.74	2.8	1.3	599
	South	South sound	69	68	2.9	16	69	4.15	3.5	2.3	747
		All marine sites	129	128	37.5	28	76	4.83	3.2	1.9	683
Total		All sites	763	688	11.8	204	53	4.82	3.7	2.3	758

along the migratory route of the Puget Sound fish. Thus, the elevated PCB levels observed for Puget Sound Chinook salmon relative to coastal populations were probably associated with differences in PCB contamination in the environments they inhabit or with differences in diet.

Freshwater versus Marine Sources of PCBs

Our results indicated that the elevated concentrations in subadult and maturing Chinook salmon from Puget Sound were accumulated during residence in marine habitats rather than riverine habitats in the region. Previous studies documented that out-migrating juvenile salmonids captured in the highly contaminated regions within Puget Sound (i.e., Duwamish River and Hylebos Waterway) contained higher concentrations of organic contaminants, including PCBs, associated with their riverine and estuarine habitats than did fish from hatcheries and reference areas (Stein et al. 1995; Collier et al. 1997; Meador et al. 2002; Johnson et al. 2007). However, our analysis indicated that adult Chinook salmon that had migrated as subyearlings from the Duwamish River, the most highly PCB-contaminated river draining into Puget Sound, accumulated the vast majority (>96%) of PCBs during their marine life history phase (Table 2), whereas there was little PCB contribution from freshwater. Moreover, most Chinook salmon from Puget Sound would accumulate far less PCBs from their freshwater habitats than the Duwamish River fish, as other rivers have less contaminated juvenile salmonids than the Duwamish River (Johnson et al. 2007).

These results are unsurprising given that typically over 99% of the final weight of Chinook salmon is

achieved in saltwater (Quinn 2005). On a per-gram basis, the average PCB concentration (ng/g) in subyearling Chinook salmon smolts migrating out of the Duwamish River was more than three times that in adults returning to the river. However, the estimated PCB body burden in adults was almost 170 times higher than that in smolts because adults weighed almost 600 times more than did smolts. Specifically, PCB levels for smolts that scored in the 95th percentile for body burden (i.e., the most contaminated smolts) on average accounted for less than 4% of the adult body burdens (range was from less than 1% to almost 14%). Our analysis was restricted to subyearling out-

TABLE 2.—Concentration of PCBs (ng/g) and body burden of PCBs (total ng/fish) in out-migrating Chinook salmon smolts and returning adults from the contaminated Duwamish River, Washington.

Variable	Smolts	Adults
Number of samples	80	34
Mean fish weight (g)	10	6,000
Whole body PCB concentration (ng/g) ^a		
Mean	170	57
95th percentile	860	88
PCB body burden (ng/fish) ^a		
Mean	2,100	350,000
95th percentile	9,200	800,000
Mean % of PCB body burden from the most contaminated smolts ^b	—	3.8

^a Values for smolts are from J. P. Meador (National Oceanic and Atmospheric Administration Fisheries, Northwest Fisheries Science Center, personal communication); values for adults were estimated from measured muscle tissue concentration using the fillet-whole-body regression (see Methods) for PCBs.

^b Contaminant data were only available for out-migrating subyearling smolts, so only samples with adults that went to sea as subyearlings were included in the analysis.

TABLE 3.—Average PCB concentration (ng/g) and body burden (total number ng/fish) by saltwater age-class for Puget Sound Chinook salmon, 1992–1996.

Variable	Number of winters spent in saltwater				
	Smolts ^a	1	2	3	4
Number of samples	—	11	72	44	7
Average sample fish length (mm)	—	620	735	810	901
Average sample fish weight (g)	10	3,338	5,306	6,986	10,028
Average fillet PCB concentration (ng/g)	—	68	56	49	27
Average fillet % lipids	2.5	6.8	4.8	6.9	6.1
Average whole-body PCB concentration (ng/g) ^b	40	74	62	54	30
PCB body burden (ng/fish)	400	260,000	340,000	390,000	280,000

^a Data on PCB concentration in Puget Sound out-migrating smolts were taken from Johnson et al. (2007) and were based on fish sampled from the Nisqually River, a typical Puget Sound river. Average concentration reported by Johnson et al. (2007) for all Pacific Northwest estuaries was 27 ng/g; PCB body burden (ng/fish) was estimated using an average smolt size of 10 g.

^b Values for adults and subadults were estimated from measured fillet-whole-body regression for PCBs (see Methods).

migrants. Although yearlings spend more time feeding in rivers, the vast majority (>98.7%) of their final adult body weight is also acquired in saltwater (based on observed weights of returning adults [present study] and a 50-g average weight of yearling smolts released from Duwamish River hatcheries [K. Dimmit, WDFW, Olympia, unpublished data]). Consequently, the majority of PCB accumulation would occur in the marine environment, regardless of juvenile life history. For example, if yearling smolts from the Duwamish River had an average PCB body burden of 21,000 ng/fish, which is 10 times the average observed for subyearling smolts, the freshwater sources of PCBs would still account for only 7% of the average body burdens observed in adult fish from the Duwamish River. Additionally, for fish that spent the same amount of time feeding in saltwater, PCB concentration differences between adult fish that out-migrated as yearlings and as subyearlings were explained by differences in adult fish size (GLM on \log_e [PCBs]; SWA2 FL: $F = 15.875$, $df = 1$, $P < 0.001$; SWA3 FL: $F = 5.76$, $df = 1$, $P < 0.02$; out-migrant type and interactions were not significant for either SWA group: $P > 0.05$). Thus, we concluded that the elevated average PCB levels observed in subadult and adult Puget Sound Chinook salmon compared with other West Coast populations were associated with differences in PCB exposure during residence in marine habitats. Moreover, the wide variation among the Puget Sound samples (10–220 ng/g), which was not observed in Chinook salmon populations outside Puget Sound (USEPA 2002; Missildine et al. 2005; Rice and Moles 2006; Cullon et al. 2009), suggests that the marine ecological factors associated with PCB concentration (i.e., marine distribution and migratory patterns and associated diet) also varied considerably among individual Puget Sound Chinook salmon.

Factors Affecting PCB Accumulation in Subadult and Maturing Chinook Salmon from Puget Sound

Surprisingly, although PCBs were mostly accumulated in the marine environment, PCB exposure for Puget Sound Chinook salmon was lowest for fish that spent the longest time in saltwater (i.e., Puget Sound and the Pacific Ocean) before their upstream spawning migration. Average PCB concentrations were highest for Chinook salmon of the SWA1 group (68 ng/g) and declined for individuals that spent additional years in saltwater (Table 3). The average PCB concentrations of SWA4 fish were more typical of concentrations reported for other West Coast Chinook salmon populations (Figure 2). These results were based on analyses of a subset of 134 samples consisting of individual fish and uniform fish age composite samples. Sampling location was not included in this analysis because not all SWA classes were represented at every sampling location.

Concentration of PCBs was positively correlated with fish FL within SWA1, SWA2, and SWA3 groups (Figure 4). Both yearling and subyearling out-migrant life history types were sampled in each of these three SWA groups, but as previously stated our results showed that PCB concentration differences between the life history types could be explained by fish size. Fish of SWA4 had uniformly low PCB concentrations regardless of their size and were only represented by subyearlings.

In total, the SWA, fish FL, and lipid content accounted for 37% of the observed variation in PCB concentration among the Puget Sound Chinook salmon samples (GLM on \log_e [PCBs]; SWA: $F = 19.047$, $df = 3$; percent lipids: $F = 15.438$, $df = 1$; FL: $F = 30.402$, $df = 1$; $P < 0.001$ in all cases). Collectively, SWA and FL accounted for 30% of the PCB variation among samples, and an additional 7% was attributed to lipids.

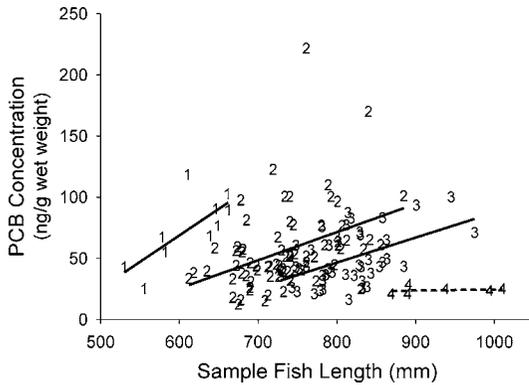


FIGURE 4.—Relationship between total PCB concentration and fish size (fork length, mm) for Chinook salmon grouped by saltwater age (SWA; number of winters [between 1 and 4] spent in saltwater). Solid lines indicate significant regressions ($P < 0.02$) for SWA1 ($r^2 = 0.53$), SWA2 ($r^2 = 0.15$), and SWA3 ($r^2 = 0.22$); the dashed line indicates a nonsignificant regression for SWA4 ($r^2 = 0.04$).

After the GLM was used to account for differences in fish size and lipid content, Puget Sound Chinook salmon that spent the longest time in saltwater (i.e., SWA4), where the majority of the PCBs are accumulated, were found to have the lowest PCB concentrations (Tukey's test: $P < 0.0001$ for all paired comparisons; Figure 5).

In addition to biological traits like SWA, sampling location might have contributed to the variation in PCB concentration among the Puget Sound samples. At each sampling location, the lowest average PCB concentrations were observed in fish that spent the most time in saltwater, which is consistent with the overall inverse relationship between SWA and PCB concentration observed for pooled Puget Sound samples. Despite this general pattern, population-specific differences in the PCB concentration within SWA classes were also observed; however, a full analysis of SWA, PCB concentration, and sampling locations was not possible because we did not sample all SWA classes at all locations. Specifically, for fish of SWA2 (the only SWA class that was well represented at most sampling locations), the Deschutes River fish in southern Puget Sound had significantly higher PCB concentrations than those from the Nooksack and Skagit rivers in northern Puget Sound and the Nisqually River in southern Puget Sound after adjusting for lipid and FL effects (GLM on $\log_e[\text{PCBs}]$; location: $P = 0.011$; FL: $P = 0.003$; percent lipids: $P = 0.014$; Tukey's test: $P < 0.05$ for all comparisons). Within the SWA2 group, sampling location accounted for 23% of the observed variation, followed by FL (8%) and percent lipid (6%).

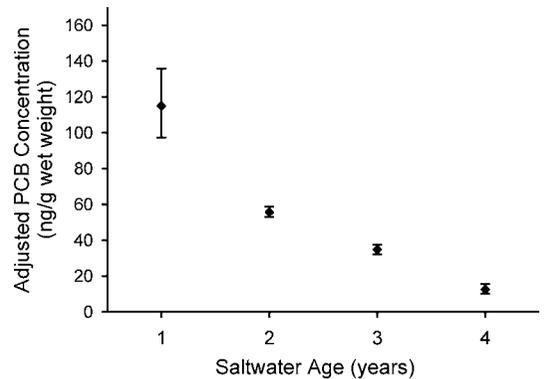


FIGURE 5.—Relationship between Chinook salmon saltwater age (number of winters [between 1 and 4] spent in saltwater) and PCB concentration (mean \pm SE) in filets. Concentrations are geometric means adjusted (by general linear model [GLM]) to grand means of fork length and percent lipids ($r^2 = 0.37$, $P < 0.0001$).

This suggests that the sampling location, in addition to biological traits, affects PCB accumulations, but additional sampling would be needed to partition the relative importance of these factors.

Switching to cleaner prey and growth dilution may partially explain the inverse relationship between SWA and PCB concentration. Prey switching and growth dilution can result in a lowering of PCB concentration (ng/g fish tissue) in older fish; however, PCB body burden (total ng/fish) should at least remain constant in older fish once the shift to cleaner prey has occurred. The low PCB concentration in SWA4 fish (Table 3) cannot be fully explained by a scenario in which SWA3-type fish simply remain in saltwater for a fourth year while shifting their diet to cleaner prey during that year. However, SWA4 Chinook salmon had lower (albeit not statistically significant) PCB body burdens than SWA2 and SWA3 fish.

Most interestingly, the average PCB body burden of Chinook salmon belonging to the SWA4 group was more similar to that of SWA1 fish (Table 3) despite three additional years of feeding and growth. One possible explanation for this pattern is that both SWA1 and SWA4 fish fed on contaminated prey through their first winter in saltwater, after which SWA1 fish returned to rivers and SWA4 fish moved to areas outside of Puget Sound, where their diets shifted to cleaner prey. Alternatively, the SWA4 fish could have fed on lightly contaminated prey throughout their time spent in the marine environment. Thus, we suggest that two mechanisms affected the PCB exposure of Puget Sound Chinook salmon: (1) growth dilution of PCB associated with the addition of weight accumulated by

older fish and (2) a reduction in dietary PCB inputs associated with feeding on cleaner prey for fish that spent the longest time in saltwater. Because SWA2 and SWA3 fish exhibited greater PCB body burdens than either SWA1 or SWA4 fish, it seems likely that their exposure to contaminated prey continued past their first year. Collectively, Chinook salmon from the four SWA groups we sampled probably represent distinct populations that experienced differential exposure to PCBs.

Furthermore, given that only 37% of the observed PCB variation was explained by biological traits (i.e., SWA, FL, and percent lipid content), other unmeasured factors are involved. We hypothesized that resident behavior by Puget Sound Chinook salmon in the contaminated waters of the sound could account for (1) the elevated PCB concentrations in these fish compared with other West Coast populations and (2) the variation in PCB levels among SWA groups. In the following section, we estimate the proportion of Puget Sound Chinook salmon that reside and feed in Puget Sound, thereby increasing their exposure to PCBs, rather than migrating to the Pacific Ocean.

Residency of Chinook Salmon in Puget Sound and Implications for PCB Exposure

Chinook salmon originating from Puget Sound typically migrate to coastal Canadian waters of the Pacific Ocean (Myers et al. 1998; Quinn 2005), but fishery managers within the region have long known that some proportion remains in Puget Sound and contributes to a year-round fishery (Smith 1920; Pressey 1953; Haw et al. 1967). Chinook salmon that migrate through or reside and feed in Puget Sound, particularly inner Puget Sound, would experience a much more contaminated environment than other populations along the West Coast of North America. Puget Sound is a deep, fjord-like estuary with a narrow connection to oceanic waters through the Strait of Juan de Fuca and shallow sills at Admiralty Inlet that tend to isolate its waters from cleaner oceanic waters (Harrison et al. 1994) and to reduce the summer flushing time relative to that of the nearby Strait of Georgia (Thomson 1994). The hydrological isolation of Puget Sound from the Pacific Ocean serves to entrain water, nutrients, and contaminants within it. Consequently, the pelagic food web in Puget Sound is more heavily contaminated than that in the coastal waters as evidenced by the relatively high PCB levels in mussels from the central and southern basins (Puget Sound Action Team 2007). Indeed, within Puget Sound, Pacific herring, which are common prey of Chinook salmon, have substantially higher PCB levels than those from the Strait of Georgia and from Alaska (West et al. 2008).

Population-specific catch estimates from the FRAM supported our hypothesis that a considerable proportion of Puget Sound Chinook salmon reside in the inland marine waters of Washington and British Columbia in winter fisheries when more migratory fish would have been distributed in coastal Pacific Ocean waters. The FRAM results revealed that at least 29% of the Puget Sound-origin Chinook salmon that migrated to saltwater as subyearlings and 45% of those that migrated as yearlings were caught in the inland marine waters of Washington and British Columbia as subadults and maturing adults in the winter months during the years of our study (Table 4). By definition, the fish caught in the winter months were displaying resident behavior; however, differences in the intensity and regulation of fisheries in coastal and Puget Sound waters made it impossible to estimate the precise proportion of Puget Sound Chinook salmon that were resident during our study. Nonetheless, results of the FRAM robustly support two important general conclusions: (1) that a substantial percentage of Puget Sound Chinook salmon fed in Puget Sound during winter months and (2) that this percentage was higher for subadult and maturing fish that out-migrated as yearling smolts than for fish that out-migrated as subyearlings. Furthermore, our estimates of the proportion of Puget Sound fish displaying resident behavior were conservative because our calculations do not include any resident fish that may be caught in Puget Sound fisheries during July through September.

In addition to documenting that a considerable proportion of Puget Sound-origin Chinook salmon displayed resident behavior, a subset of these fish were caught in inner Puget Sound, the more highly urbanized basins in the study area. From 1992 to 1996, almost 12% of the total catch of Puget Sound-origin subadults and maturing adults that migrated to saltwater as subyearlings and 20% of the total catch of those that out-migrated as yearlings were caught in inner Puget Sound (Table 4).

The FRAM results also revealed that for both juvenile out-migrant life history types, a higher proportion of younger fish than older fish were caught in the winter months (Table 5). Among Puget Sound-origin Chinook salmon that migrated to saltwater as subyearlings, the Puget Sound winter fisheries took 16% of the total catch of 3-year-olds but only 5% of the total catch of 5-year-olds. Higher catches of younger fish in inland winter fisheries could mean that resident fish mature at an earlier age or that younger fish reside in Puget Sound for a few years before migrating to the Pacific Ocean. Perhaps more importantly, Chinook salmon that migrated to the coastal Pacific Ocean would only have been sampled or caught in Puget

TABLE 4.—Estimated percent (%) of Chinook salmon originating from Puget Sound caught in the inland marine waters of Washington (WA) and British Columbia (BC) and in inner Puget Sound during the nonmigratory period. Data in this table were generated from the Fishery Regulation Assessment Model, a simulation model based on tagging data that is used to assess fishery impacts (see text for details).

Release age	Capture year	Number of fish landed ^a	Percent caught during nonmigratory period ^b	
			Inland marine waters of WA and BC	Inner Puget Sound ^c
Subyearling	1992	164,755	35.0	7.4
	1993	129,719	25.0	9.7
	1994	114,109	19.5	7.9
	1995	109,543	32.2	18.4
	1996	102,613	31.9	17.3
	Average		28.7	12.1
	Yearling	1992	31,215	38.3
1993		25,596	44.7	19.9
1994		26,941	33.6	17.3
1995		25,478	54.9	26.3
1996		16,394	56.1	20.0
Average			45.5	20.1

^a Catch areas include Alaska; northern coast of BC; west coast of Vancouver Island; and WA, Oregon, and California coasts.

^b Percent caught during the nonmigratory period was estimated as percent of total Puget Sound-origin fish landed in fisheries that occurred during January–June and October–December outside the typical migration timing for adults returning to spawn (July–September). This is a conservative estimate of the number of fish displaying resident behavior because we assumed all other fish caught inside Puget Sound, the Strait of Georgia, and the Strait of Juan de Fuca during July–September fisheries and those caught outside these areas during the remainder of the year had a more oceanic marine distribution.

^c Percent of fish caught in inner Puget Sound (i.e., south of Admiralty Inlet) is a subset of the total number of fish caught in the inland marine waters.

Sound fisheries during the year when they were returning to spawn, whereas the residents would have been available to sampling and to the fishery during previous years. These explanations are not mutually exclusive but in all cases are consistent with the idea that the younger sampled fish were more likely to have been resident for a substantial portion of their lives.

Feeding in the contaminated Puget Sound environment probably contributes to the elevated PCB concentrations of these Chinook salmon compared

with other West Coast populations. Notwithstanding the differences in habitat between Puget Sound and the West Coast, the prey taxa and general diet of Puget Sound resident Chinook salmon are similar to those reported for Chinook salmon elsewhere. Higgs et al. (1995) documented that in marine waters, Chinook salmon generally eat a mixture of crustaceans, squid, and fish, especially Pacific herring and Pacific sand lance *Ammodytes hexapterus*, as they grow. Historical studies on diet of Puget Sound resident Chinook

TABLE 5.—Estimated percentage (%) of Chinook salmon originating from Puget Sound within each juvenile life history type (migrating as subyearlings or yearlings) and age-class caught in inner Puget Sound outside the usual migration window for adult fall-run Chinook salmon, 1992–1996^a.

Catch year	Subyearling age-class (years) ^b			Yearling age-class (years) ^b		
	3	4	5	3	4	5
1992	12.5	6.8	5.2	17.4	18.6	4.6
1993	13.2	4.9	6.0	20.8	22.0	3.9
1994	14.6	7.2	6.0	17.6	17.5	8.5
1995	25.1	10.8	4.2	26.1	28.7	9.6
1996	16.4	19.2	3.4	17.5	24.9	6.5
Average	16.4	9.8	5.0	19.9	22.3	6.6

^a All 3–5-year-old fish caught in inner Puget Sound (catch areas 9, 10, 11, and 13) outside the typical (July–September) migration time for fall-run adults returning to Puget Sound were presumed to be resident.

^b Two-year-old fish were excluded from this analysis because most were too small to recruit to fisheries outside the typical (July–September) migration time for adults returning to Puget Sound. Less than 5% of 2-year-olds released as subyearlings and 0% released as yearlings were caught in this time frame.

salmon also documented that fish, especially Pacific herring, formed the majority of their diet by volume, followed by crustaceans and squid (Kirkness 1948).

Marine-caught fish from central Puget Sound had a high probability of being residents based on their demographics and also had some of the highest PCB levels (Figure 3). These fish were shorter, were younger in total age, and had spent fewer winters in saltwater than fish from the other marine sampling location and fish returning to Puget Sound rivers (Table 1). The FL of sampled fish varied significantly among locations (ANOVA on $\log_e[FL]$: $P < 0.001$), and overall the central Puget Sound fish were the shortest fish sampled (Table 1; Tukey's test: $P < 0.001$ for all comparisons). Seventy-seven percent of the 60 fish contributing to the 12 central Puget Sound composite samples belonged to the SWA1 group compared with 0–13% at other locations, suggesting that these fish were immature residents. Moreover, the central Puget Sound sampling location is near Apple Cove Point, an area that is well known to sport fishers for its abundance of resident Chinook salmon. Whether fish resided in their area of capture for extended periods is unknown. However, fish residing and feeding in central and southern Puget Sound probably would be exposed to higher PCB levels than fish feeding in northern areas of the sound. Pacific herring collected from central Puget Sound are more highly contaminated with PCBs than those from northern Puget Sound and the southern Strait of Georgia (West et al. 2008).

Overall, we conclude that the wide range of PCB levels observed for Puget Sound Chinook salmon reflects their degree of residency and distribution while feeding in marine waters. Unfortunately, the amount of time for which Chinook salmon reside in Puget Sound and the distribution of individual fish are unknown. Brannon and Setter (1989) proposed that some Chinook salmon within a population overwinter in Puget Sound but make short annual migrations from inner Puget Sound to the Strait of Georgia and back until they reach maturity or are caught. Historical tagging studies also suggested that some southern Puget Sound populations were more resident as they were over-represented in winter fisheries relative to northern Puget Sound populations (Haw et al. 1967).

Although hatchery practices (e.g., size at release and date of release) in the Puget Sound region have been manipulated to encourage residency so that local rather than distant fisheries receive the benefit of the hatchery's production (Appleby and Doty 1995), residency is probably a natural phenomenon in Puget Sound Chinook salmon. Most of the Puget Sound-origin Chinook salmon that were caught between 1992 and 1996 originated from hatcheries. However, in past

decades, resident Chinook salmon were taken in recreational fisheries when the populations were predominantly wild (Pressey 1953). More resident populations of Chinook salmon have also been recorded for British Columbian populations whose natal streams are located on the east side of Vancouver Island in the Strait of Georgia (Healey and Groot 1987), suggesting that resident Chinook salmon populations with more localized marine distributions may exist along many areas of the West Coast that have protected, fjord-like waters.

In aquatic ecosystems elsewhere, elevated PCB concentrations in salmon are only found in populations whose movements are confined to highly polluted bodies of water. Atlantic salmon *Salmo salar* from the Baltic Sea have elevated PCB concentrations associated with their residency within contaminated inland marine and estuarine waters. The PCB concentrations in Baltic Sea Atlantic salmon were approximately five to six times higher than concentrations observed during our study (Berghlund et al. 2001; Isosaari et al. 2006). However, unlike Puget Sound (where only some Chinook salmon are resident), almost all (>99.5%) Baltic Sea Atlantic salmon reside within the semi-enclosed Baltic Sea during their marine phase (Karlsson and Karlström 1994), when they feed on PCB-contaminated prey like European sprat *Sprattus sprattus* and Atlantic herring *C. harengus* (Berghlund et al. 2001). The most contaminated Puget Sound Chinook salmon, which we believe resided in Puget Sound until they reached maturity, had concentrations comparable with those observed in Baltic Sea Atlantic salmon. Higher concentrations of PCBs in North American salmon are only observed in the Great Lakes (Miller 1994; Stow et al. 1994), where Chinook salmon and coho salmon populations in the early 1990s were 20–30 times more contaminated with PCBs than the Puget Sound Chinook salmon we studied.

Implications of Elevated PCB Contamination in Resident Chinook Salmon

The elevated PCB levels associated with the residency of Chinook salmon in Puget Sound have resulted in consumption advisories and also have implications for the viability of these fish and the southern resident killer whales that feed upon them. The Washington Department of Health (2006) recommended that people limit their consumption of Puget Sound Chinook salmon to 1 meal/week based on an evaluation of PCB levels in fish samples collected for this study. Sport fishers targeting resident Chinook salmon in winter fisheries were also advised to limit consumption of these fish to no more than 2 meals/

month because of the significantly higher PCB concentrations.

The health of some Puget Sound Chinook salmon may be adversely affected because their PCB concentrations exceeded the threshold levels of PCBs known to cause adverse effects in salmonids. Almost 22% of the maturing and subadult Chinook salmon samples we collected from Puget Sound had PCB concentrations above an effects threshold identified for salmonid fishes (i.e., 2,400 ng/g lipids), which included endpoints such as reduced growth, altered enzyme and hormone levels, and increased mortality (Meador et al. 2002). This threshold was based on whole-body concentrations rather than fillet concentrations; therefore, further investigations on whole-body samples are needed to evaluate the extent to which the health of subadult and maturing Chinook salmon from Puget Sound is impaired. Moreover, due to the biomagnification of PCBs in long-lived predatory species, the observed average PCB levels in Puget Sound Chinook salmon (53 ng/g) may not protect the health of southern resident killer whales that feed in Puget Sound. Based on modeled PCB values for whales, Hickie et al. (2007) concluded that a diet at the tissue residue guideline of 50 ng/g, similar to the average concentration in Puget Sound Chinook salmon, would place over 95% of the killer whale population above a PCB effects threshold of 17 mg/kg in blubber; this threshold has been associated with immune function and endocrine endpoints in harbor seals *Phoca vitulina*.

Although Puget Sound Chinook salmon have elevated PCB levels compared with other West Coast populations, lower concentrations are expected for the other four Pacific salmon species that originate from the region. The concentration of PCBs will reflect among-species differences in marine distribution (and resulting proximity to contaminated prey), diet, duration of exposure (i.e., life span), and lipid content, factors that are not independent of each other (O'Neill et al. 1998). Overall, Chinook salmon and coho salmon have a more coastal marine distribution along the continental shelf than do sockeye salmon *O. nerka*, pink salmon *O. gorbuscha*, and chum salmon *O. keta* (Quinn 2005); therefore, Chinook salmon and coho salmon can be more readily exposed to contaminants that are present in coastal waters. Sockeye salmon, pink salmon, and chum salmon seldom reside in Puget Sound but instead migrate to cleaner environments of the Pacific Ocean to feed. Coho salmon also reside in Puget Sound, but they feed in marine waters for only a year and a half and typically feed at a lower trophic level than do Chinook salmon. The proportion of fish in the diet is greatest for Chinook salmon, followed by coho salmon, pink salmon, sockeye salmon, and chum

salmon (Fresh et al. 1981; Peterson et al. 1982; Beacham 1986; Higgs et al. 1995); the greater extent of piscivory in Chinook salmon results in a longer food chain and potentially greater contaminant exposure. Previous studies in the Puget Sound region have shown that average PCB levels are higher in Chinook salmon than in coho salmon (O'Neill et al. 1998) and that PCB levels are lowest in sockeye salmon (McIntyre and Beauchamp 2007).

Finally, it is important to note that the PCB levels for Puget Sound Chinook salmon and coho salmon were probably higher in the 1970s and 1980s than at present (West and O'Neill 2007). Historical data are limited, but PCB levels in Puget Sound coho salmon appear to have declined rapidly during the 1970s through the mid-1980s, whereas the decline has slowed or leveled off more recently (Stout and Beezhold 1981; USEPA 2002), which is consistent with national and global trends for fish throughout the northern hemisphere (Matta et al. 1986; Stow et al. 1994; Bignert et al. 1998; Lamon et al. 1999; Hickey et al. 2006). Current PCB levels in Puget Sound Chinook salmon probably represent both historical and ongoing loadings of these contaminants into Puget Sound (e.g., continued leakage from upland contaminated sites and long-range atmospheric transport from countries where PCBs are still in use) and cycling among environmental compartments. Given these trends in biota and the estimated rates of degradation and burial of PCBs in geological reserves (Jönsson et al. 2003), PCB levels in Puget Sound Chinook salmon and coho salmon are unlikely to decline substantially in the near future unless substantial reductions in loadings to Puget Sound are made.

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