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# Persistent Organic Pollutants in Juvenile Chinook Salmon in the Columbia River Basin: Implications for Stock Recovery

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## ARTICLE

## Persistent Organic Pollutants in Juvenile Chinook Salmon in the Columbia River Basin: Implications for Stock Recovery

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#### Abstract

Among the populations of Pacific salmon and steelhead *Oncorhynchus mykiss* (anadromous Rainbow Trout) that inhabit the Columbia River basin there are currently 13 Evolutionarily Significant Units listed as threatened or endangered under the U.S. Endangered Species Act. While habitat loss, dams, overharvest, and climate change have been implicated in declining abundance of Chinook Salmon *O. tshawytscha* in the Columbia River, chemical contaminants represent an additional, yet poorly understood, conservation threat. In this study we measured concentrations of persistent organic pollutants in juvenile Chinook Salmon from various Columbia River stocks and life history types to evaluate the potential for adverse effects in these threatened and endangered fish. Polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane (DDTs), recognized contaminants of concern in the Columbia basin, are the primary focus of this paper; other contaminants found in these fish, such as polybrominated diphenyl ethers and polycyclic aromatic hydrocarbons, are described in other publications. We frequently detected PCBs and DDTs in juvenile salmon and salmon diet samples from the lower Columbia River and estuary. In some cases, concentrations in salmon were above estimated thresholds for effects on growth and survival. The tidal freshwater portion of the estuary, between Portland, Oregon, and Longview, Washington, appeared to be an important source of contaminants for juvenile salmon and a region in which salmon were exposed to toxicants associated with urban development and industrial activity. Highest concentrations of PCBs were found in fall Chinook Salmon stocks with subyearling life histories, including populations from the upper Columbia and Snake rivers, which feed and rear in the tidal freshwater and estuarine portions of the river for extended periods. Spring Chinook Salmon stocks with yearling life histories that migrate more rapidly through the estuary generally had low PCB concentrations, but high concentrations of DDTs. Lipid content was low (<1%) in many of the fish examined, contributing to high lipid-adjusted contaminants concentrations in some samples.

Thirteen evolutionarily significant units (ESUs; Waples 1991; NRC 1996) of Pacific salmon and steelhead Oncorhynchus mykiss (anadromous Rainbow Trout) that rear and spawn in the Columbia River basin are recognized as threatened or endangered under the U.S. Endangered Species Act (ESA; Good et al. 2005). Multiple factors, including overharvest, the stress of dam passage and other impacts of the Columbia River hydropower system, climate change, predation, and loss and alteration of both estuarine and freshwater habitats have contributed to their decline (NRC 1996; Fresh et al. 2005; Williams et al. 2005, Kostow 2009, NMFS 2010; Bottom et al. 2005, Sheer and Steel 2006; Honea et al. 2009; Roegner et al. 2010). Chemical contaminants are an additional, yet poorly understood, conservation threat to Columbia River Pacific salmon, especially for stocks with longer residence times in the heavily populated lower Columbia River and estuary (LCREP 2007; USEPA 2009; ISAB 2011).

In previous studies we have established that juvenile Chinook Salmon *O. tshawytscha* in the Columbia River are absorbing a variety of contaminants, including polychlorinated biphenyls (PCBs); dichloro-diphenyl-trichloroethanes (DDTs), and other organochlorine pesticides; the flame retardants, polybrominated diphenyl ethers (PBDEs); polycyclic aromatic hydrocarbons (PAHs); various current-use pesticides, and estrogenic compounds (Johnson et al. 2007a, 2007b; LCREP 2007; Sloan et al. 2010; Yanagida et al. 2012, Morace 2006). Exposure to these chemicals may lead to altered immune function and increased disease susceptibility (Arkoosh et al. 1994, 1998, 2001; Bravo et al. 2011), poor growth, or metabolic dysfunction (Meador et al. 2002, 2006) and altered behavior (Scholz et al. 2000; Sandahl et al. 2005), all of which increase the risk of mortality in juvenile salmon.

Columbia River Chinook Salmon, however, are composed of multiple stocks and life history types with different geographic ranges, migration timing, and length of freshwater and estuary residence (Fresh et al. 2005). While we know that juvenile Chinook Salmon are accumulating chemical contaminants, we know much less about the routes of exposure for different Chinook Salmon stocks, the types of contaminants that various life history types and stocks are accumulating, and potential impacts of these contaminants on the health of individual fish and recovery of listed stocks.

Our study objectives were to document concentrations and examine spatial trends of PCBs and DDTs in out-migrant Columbia River juvenile Chinook Salmon, and in particular, to examine the influence of life history type and stock of origin on PCB and DDT exposure profiles and the associated risk of toxic injury to threatened and endangered Pacific salmon. We expanded the geographical range of our initial study performed with juvenile Chinook Salmon in 2001 and 2002 (Johnson et al. 2007b) to include out-migrant juvenile Chinook Salmon from the eight ESUs present in the Columbia Basin (Myers et al. 1998), which we sampled from thirteen sites in the lower Columbia and lower Willamette rivers. This region provides rearing and spawning habitat for fish of lower Columbia River Chinook and upper Willamette River spring Chinook ESUs. The region is also used as a migration corridor for all Columbia River Chinook Salmon stocks, and as rearing habitat for fall-run Chinook Salmon stocks from the interior Columbia River basin (Myers et al. 1998).

We chose to focus on PCBs and DDTs for this study because they represent chemical classes associated with two different land use patterns, PCBs being associated with historical urban and industrial activities and DDTs being associated with historical agrochemical use. Also, threshold concentrations of PCBs and DDTs associated with toxic effects on Pacific salmon have been estimated (Meador et al. 2002; Beckvar et al. 2005), so their likely impacts on Pacific salmon health and survival can be more readily evaluated. Data on exposure to PBDEs and PAHs in juvenile Columbia River Chinook Salmon from the same study area are available in Sloan et al. (2010) and Yanagida et al. (2012) but are not included in this analysis.

We hypothesized that contaminant accumulation in juvenile salmon would be influenced by their life history type and particular ESU, their geographical distribution, and the land use patterns in the areas where they occur, and so, we here provide a brief overview of these factors.

# CHINOOK SALMON LIFE HISTORY TYPES, ESUS, AND GEOGRAPHIC RANGES

## Life History Types

Columbia River juvenile Chinook Salmon can generally be classified into one of two major life history types, subyearlings and yearlings, based on age at emigration from freshwater (Table 1). Yearlings spend their first year in tributaries and downstream freshwater rearing habitats before migrating to sea the following spring. Yearlings spend minimal time in the Columbia River estuary (Brannon 2004; Fresh et al. 2005). In contrast to yearlings, subyearlings migrate to the ocean during their first year as fry or smolts and may spend up to several months rearing in the estuary before entering the ocean (Dauble and

TABLE 1. Chinook Salmon Evolutionarily Significant Units (ESUs), juvenile life history types, and genetic stock groups in the Columbia River basin. Minor juvenile life history type is indicated in parenthesis. Life history information is from Waples et al. (2004). The genetic stock groups given are those used in our analysis.

ESU and ocean-entry type	Genetic stock group
Lower Columbia River	
Subyearling, <sup>a</sup> yearling <sup>b</sup>	West Cascade tributary fall, West Cascade tributary spring, and Spring Creek Group fall
Upper Willamette River <sup>c</sup>	
Yearling (subyearling)	Willamette River spring
Deschutes River summer-fall	
Subyearling (yearling)	Deschutes River summer/fall
Mid Columbia spring	
Yearling	Mid and upper Columbia River spring
Upper Columbia Summer–Fall	
Subyearling (yearling)	Upper Columbia River Summer/Fall
Upper Columbia River Spring	
Yearling	Mid and upper Columbia River spring
Snake River Fall	
Subyearling (Yearling)	Snake River fall
Snake River Spring–Summer	
Yearling	Snake River spring

<sup>a</sup>Fall run Chinook Salmon in the lower Columbia River ESU are subyearling type. <sup>b</sup>Spring run Chinook Salmon in the lower Columbia River ESU are yearling type. <sup>c</sup>Chinook Salmon in the upper Willamette River ESU are spring run.

Watson 1997; Fresh et al. 2005). Yearlings and subyearlings also differ in their patterns of habitat use as they migrate through the lower river and estuary. The larger yearling migrants typically utilize deeper main-stem channels, whereas the smaller subyearling out-migrants use peripheral tidal marshes, shallow side channels, and forested marsh habitats for rearing (Fresh et al. 2005).

## **Columbia River Chinook Salmon ESUs**

Chinook Salmon populations in the Columbia River basin have been grouped into several discrete ESUs based on a synthesis of genetic, life history, geographic, and environmental data (Myers et al. 1998). Chinook Salmon ESUs in this basin are described below; their ranges and characteristics are also summarized in Table 2 and mapped in Figure 1. Geographic ranges are described in terms of Columbia Basin Ecological Provinces and subbasins within those provinces (CBFWA 2008).

Lower Columbia River ESU.—Chinook Salmon from this ESU occupy Columbia River tributaries from the mouth of the Columbia to the Klickitat River, in the Columbia Gorge and lower Columbia Basin provinces (CBFWA 2008). In our analysis, this ESU contains three genetic groups of Chinook Salmon: Spring Creek Group fall, West Cascade tributary spring, and West Cascades tributary fall. The Spring Creek Group fall stock consists of populations that are genetically similar to fall Chinook Salmon from the Spring Creek National Fish Hatchery, which have been out-planted extensively throughout the ESU (Myers et al. 2006). Contemporary sources of Spring Creek Group fall stock include several tributaries in the Columbia Gorge as well as in the lower river (Myers et al. 2006; Smith and Engle 2011). The West Cascades spring and fall run stock groups originate primarily in tributaries of the Cowlitz, Kalama, Lewis, and Sandy river subbasins.

Upper Willamette River ESU.—This ESU contains naturally spawning populations of spring Chinook Salmon originating in the Willamette River subbasin of the lower Columbia River Estuary province. Populations in this ESU have complex juvenile life history patterns including both yearling and subyearling migrants (Myers et al. 2006). Willamette River hatchery spring Chinook Salmon have also been released outside of the historic range of the ESU, most notably in the Sandy River (Myers et al. 2006). As a result of these releases, spring Chinook originating in the Sandy River are genetically similar to those in the Willamette River and may potentially contribute to our estimates of that stock in our juvenile catches.

Upper Columbia River Summer–Fall ESU.—This ESU contains Chinook Salmon spawning in tributaries to the Columbia River in the Columbia Cascades and portions of the Columbia Plateau provinces, from the confluence of the Snake and Columbia rivers upstream to the Chief Joseph Dam. Most naturally produced juveniles in the ESU are subyearling migrants (Waples et al. 2004). The largest source of naturally produced fall-run fish is from spawners in main-stem Columbia River habitats in the Hanford Reach area. Chinook Salmon populations derived from fall run fish in the ESU are also currently found downstream of the ESU's historical range, in the Columbia River gorge (Smith and Engle 2011) and in main-stem spawning habitats below Bonneville Dam (Myers et al. 2006).

Snake River Fall ESU.—This ESU includes all native populations of fall-run Chinook Salmon in the main-stem Snake River as well as the Tucannon, Grand Ronde, Imnaha, Salmon, and Clearwater river subbasins. This region is located within the Blue Mountain, Mountain Snake, and Columbia Plateau provinces.

Snake River Spring–Summer ESU.—This ESU includes all natural populations of spring-run, Chinook Salmon in the mainstem Snake River and the Tucannon, Grand Ronde, Imnaha, and Salmon river subbasins. Juveniles in this ESU are thought to be entirely yearling migrants (Waples et al. 2004). The majority of the spawning habitat occurs in the northeast part of the Columbia Basin, in the Blue Mountain, and Mountain Snake provinces.

Deschutes River Summer–Fall ESU.—This ESU was initially included in the Snake River Fall Chinook Salmon ESU (Myers et al. 1998) but was subsequently designated as a separate ESU (WCCSBRT 1999). This ESU includes all naturally spawning populations of summer-run and fall-run Chinook Salmon in

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TABLE 2. Description of Columbia River Chinook Salmon by Evolutionarily Significant Unit (ESU) as defined by NMFS (Myers et al. 1998, 2006; ICTRT 2003) and, in parentheses, their listing status under the U.S. Endangered Species Act. Ecological provinces and subbasins are as defined in CBFWA (2008).

	Land uses in areas of					
Geographic origin	occurrence	Contaminant types				
ESU: lower Col	umbia Chinook Salmon (threa	tened, 1999)				
Columbia Gorge, lower Columbia and Columbia estuary Provinces; Columbia River tributaries from the mouth of the Columbia to the Klickitat River	Forestry, grazing; agriculture and urban/industrial	Current use and legacy pesticides, industrial contaminants, wastewater compounds				
FSU: unner Will	amette Chinook Salmon (three	atened 1999)				
Lower Columbia-Willamette subbasins; Willamette River basin above the Willamette Falls.	Agriculture; uban and industrial uses	Current use and legacy pesticides, industrial contaminants, wastewater compounds				
ESU: Snake Riv	er fall Chinook Salmon (threa	tened, 1999)				
Mountain Snake and Columbia Plateau provinces; main-stem Snake River and the Tucannon River, Grande Ronde River, Imnaha River, Salmon River, and Clearwater River subbasins.	Rangeland, agriculture; also urban development in the southern half of Idaho.	Current use and legacy pesticides, industrial contaminants, wastewater compounds)				
ESU: Snake River spr	ing-summer Chinook Salmon	(threatened, 1999)				
Mountain Snake province; tributaries to the main-stem Snake River, including Tucannon River, Grande Ronde River, Imnaha River, and Salmon Rivers subbasins.	Forestry, some mining and grazing.	Current use and legacy pesticides, metals				
ESU: upper Columbia	<b>River spring Chinook Salmon</b>	(endangered, 1999)				
Columbia Cascades province; Columbia River tributaries upstream of the Rock Island Dam and downstream of Chief Joseph Dam in Washington State (Wenatchee, Entiat, and Methow River subbasins).	Forestry, grazing, some agriculture	Current use and legacy pesticides				
Upper Columbia River summer-fall Chinook Salmon (Not warranted)						
Columbia Cascades province; Columbia River basin from Yakima River to the U.S.–Canada border (Yakima. Wenatchee, Entiat, Methow, and Okanogan River Subbasins.	Forestry, grazing, some agriculture	Current use and legacy pesticides; industrial contaminants during out-migration				
Deschutes sum	ner–fall Chinook Salmon (Not	warranted)				
Columbia Plateau province, Deschutes River basin	Forestry, grazing, some agriculture	Current use and legacy pesticides; industrial contaminants during out-migration				
Middle Columbi	a spring Chinook Salmon (No	t warranted)				
Columbia Plateau Province; tributaries to the Columbia River from the Klickitat River basin upstream to the Yakima River basin (i.e., the Klickitat, Deschutes, John Day, and Yakima rivers).	Agriculture, grazing, mining	Nitrates, sulfites, and pesticides, heavy metals				

the Deschutes River basin. Nonpassable dams placed on the Deschutes River have eliminated anadromous runs of Chinook Salmon into the upper Deschutes River basin. These fish are not currently listed under the ESA.

## Land-Use Patterns in Chinook Salmon Critical Habitat

The land uses and contaminants of concern within Chinook Salmon critical habitat (see Table 2), are described in detail elsewhere (LCREP 2007; CBFWA 2008; NMFS 2008,



FIGURE 1. Distribution of Chinook Salmon evolutionarily significant units in the Columbia River basin (CBFWA 2008). [Figure available online in color.]

2009; USEPA 2009). Briefly, much of the spawning and rearing habitat and migration corridor for Columbia basin Chinook Salmon stocks lies within nonurban areas, where primary land uses are forestry, livestock grazing, and agriculture (CBFWA 2008). This is the case for the Mountain, Snake, and Blue Mountain provinces (provide spawning and rearing areas for the Snake River fall and Snake River Spring-Summer ESUs), the Columbia Plateau (provides spawning and rearing habitat for the Deschutes River Summer-Fall ESU, upper Columbia River Summer-Fall ESU, and the mid and upper Columbia River Spring ESUs), the Columbia Cascades Province (contains spawning habitat for the mid and upper Columbia spring Chinook Salmon), and the Columbia Gorge province (provides habitat for lower Columbia Spring Creek Group fall Chinook Salmon and upper Columbia summer-fall Chinook Salmon). While there are also inputs of industrial contaminants within these regions, particularly in the Columbia Plateau near Hanford Reach and the Tri-Cities (USEPA 2009), forestry and agriculture are more dominant land uses.

In contrast, the Lower Columbia Province, which provides spawning and rearing area for Willamette River and lower Columbia River ESU, as well as rearing habitat for Interior Columbia basin fall Chinook Salmon stocks, contains multiple human population centers, including the three largest cities in Oregon (Portland, Salem, and Eugene/Springfield) and the fourth largest in Washington (Vancouver). While this area constitutes a small percentage of overall acreage, it has the greatest population density within the Columbia River basin, and therefore a major impact on water quality. The majority of wastewater discharges, as well as point-source pollutant discharges from industry, originate in the region (USEPA 2009). Nonurban areas in the Lower Columbia Province are used primarily for forestry and agriculture (CBFWA 2008),

## Hypotheses on Chinook Salmon Contaminant Exposure and Risk

Based on land-use and contaminant distributions in the Columbia basin, and the ranges and habitat-use patterns of the different Chinook Salmon life history types and stocks that are present in the region, we hypothesized that subyearling fish (i.e., Snake River fall, upper Columbia summer-fall, Spring Creek Group fall, and West Cascades fall) would generally have higher body burdens of and greater risk of toxic injury from PCBs than yearling fish because of their more extended rearing period in the lower Columbia River, where industrialization and urbanization are most extensive. The risk to yearling fish (i.e., Snake River and Upper Columbia River spring Chinook Salmon) would be primarily from exposure to DDTs and other agricultural pesticides, which are more prevalent in their spawning and rearing habitat. We also anticipated that Willamette River spring Chinook Salmon would show substantial accumulation of both contaminant-classes because they would probably be rearing in areas with agricultural and urban land uses. Additionally, we hypothesized that West Cascades fall Chinook Salmon would have lower contaminant concentrations than Spring Creek Group fall Chinook salmon because many of these fish would enter the river below the major urban centers of Portland and Vancouver. In our study, we evaluate these hypotheses via the results from our field surveys of contaminant uptake in juvenile salmon from the lower Columbia River and estuary.

## **METHODS**

### **Fish and Sample Collection**

Columbia River subyearling fall Chinook Salmon were collected by beach seine from shallow-water nearshore sites (Figure 2; Table 3) on a monthly basis from April through September 2005–2009, following protocols described in Roegner et al. (2009). Juveniles less than 100 mm fork length were considered subyearlings (Fresh et al. 2005).

From 15 to 40 individuals were collected for necropsy at each site at each sampling time; all were examined for the presence of fin clips or coded wire tags that denote hatchery origin. To focus our study on naturally produced juveniles, only fish without hatchery markings were included in our study. However, because not all Chinook Salmon released from hatcheries are marked,



FIGURE 2. Locations of Columbia River sampling sites (squares) where Chinook Salmon samples were collected.

TABLE 3. Sites where juvenile salmon were sampled and samples collected in the lower Columbia River (CR) and estuary, including the Willamette River (WR). Sample type abbreviations are wb = whole body, sc = stomach contents for chemistry, g = fin clip for genetic stock identification, and dd is decimal degrees.

Site description	River kilometer	Latitude (dd)	Longitude (dd)	Years sampled	Sample type
Warrendale	CR 235	45.61250	122.026389	2005	wb, sc, g
Pierce Island	CR 229	45.620967	122.010800	2008	wb, g
Franz Lake	CR 222	45.600583	122.103067	2008, 2009	wb, g
Mirror Lake	CR 208	45.543433	122.247967	2008, 2009	wb, g
Confluence WA	CR 163	45.640833	122.718889	2005, 2008	wb, sc, g
Confluence OR	CR 163	45.673267	122.775617	2008	wb, g
Morrison Street Bridge	WR 21	45.518611	122.666667	2005	wb, sc, g
Campbell Slough	CR 145	45.783867	122.754850	2007-2009	wb, sc, g
Columbia City	CR 132	46.165967	122.94510	2005	wb, sc, g
Sandy Island	CR 121	46.015000	122.868333	2007	wb, sc, g
Lord/Walker Island	CR 100	46.137216	123.040278	2009	wb, g
Beaver Army Terminal	CR 87	46.181944	123.180556	2005	wb, sc, g
White Island	CR 72	46.159350	123.340133	2009	wb, g
Ryan Island	CR 61	46.206600	123.414817	2009	wb, g
McGowan	CR 18	46.236100	123.895600	2007	wb, g
Trestle Bay	CR 11	46.216950	123.966500	2007	wb, g
Point Adams	CR 11	46.201667	123.944444	2005	wb, sc, g
West Sand Island	CR 6	46.267950	124.005467	2005	wb, sc

our samples likely included some proportion of hatchery fish. Chinook salmon were measured (mm) and weighed (0.1 g), then sacrificed with a lethal dose of the anesthetic MS-222. The following samples were collected from the fish: stomach contents for measurement of persistent organic pollutants (POPs, including PCBs, DDTs, and various other organochlorine pesticides), bodies with stomach contents removed for measurement of lipids and bioaccumulative POPs, and fin clips were preserved in ethanol for subsequent genetic stock identification. Stomach contents for chemical analyses were removed from the gut of necropsied fish and composited by site and collection date into samples containing stomach contents from 10 to 15 individual fish each. Samples for chemical analyses were frozen and stored at  $-80^{\circ}$ C until analyzed. Samples for taxonomic analyses were preserved in 10% neutral buffered formalin.

To provide information on contaminant exposure, we also obtained whole bodies of 10 juveniles collected by purse seine in May 2007, at two deepwater sites, McCowan and Trestle Bay, in the saltwater portion of the lower Columbia River estuary (Table 3; Figure 3). These fish were processed as outline above, except that stomach contents samples were not collected. The 10 fish collected with purse seine were considered to be yearling migrants based on fork length (range, 134–160 mm).

#### Sample Analyses

*Genetic analyses.*—Genetic stock identification (GSI) techniques (see Manel et al. 2005) were used to investigate the origins of juvenile Chinook Salmon as described in Teel et al. 2009 and Roegner et al. (2010). Stock origins were

estimated using a baseline of standardized microsatellite DNA data (Seeb et al. 2007) collected from spawning populations from throughout the Columbia River basin (described in Teel et al. 2009). The GSI computer program ONCOR (Kalinowski et al. 2007), which uses the likelihood model of Rannala and Mountain (1997), was used to assign individuals to the regional Chinook Salmon genetic stock groups listed in Table 1 (Seeb et al. 2007; Teel et al. 2009).

*Lipid determination.*—We determined lipid content in Chinook Salmon whole bodies. Lipid content can be a useful indicator of salmon health (Biro et al. 2004), and also affects contaminant uptake and toxicity (Elskus et al. 2005). The tissue concentration of a lipophilic chemical that causes a toxic response is directly related to the amount of lipid in an organism (Lassiter and Hallam, 1990; van Wezef et al. 1995).

Prior to analyses, body samples from subyearling Chinook Salmon (including internal organs but without stomach contents) were composited by genetic reporting group and date and site of collection into a set of composite samples, each containing two to five fish. Larger yearling Chinook Salmon were analyzed as individuals. In Chinook Salmon body composite samples, the total amount of extractable lipid (percent lipid) was determined by thin-layer chromatography with flame ionization detection, as described in Ylitalo et al. (2005).

Chemical contaminants in whole bodies and stomach contents.—Chinook Salmon bodies and stomach contents samples, composited as described above, were extracted with dichloromethane, using an accelerated solvent extractor. The sample extracts were precleaned on a gravity-flow column



FIGURE 3. Genetic stock composition of juvenile Chinook Salmon used for chemical analysis at sampling sites in the lower Columbia River and estuary. Sites are arranged east to west from the upper limit of the lower Columba River and estuary to the mouth of the Columbia. Sites from Mirror Lake to Warrendale are considered within the Columbia Gorge; those from Trestle Bay to Morrison Street Bridge are considered below the Columbia Gorge. Abbreviations: SnakeSp = Snake River spring Chinook Salmon, UCRSp = upper Columbia spring Chinook Salmon, WRSp = Willamette River spring Chinook Salmon, LCR-WCF = lower Columbia River ESU-west Cascades fall Chinook Salmon, LCR-SCGF = lower Columbia River ESU-Spring Creek Group fall Chinook Salmon, SnakeF = Snake River fall Chinook Salmon, DeschutesF = Deschutes fall Chinook Salmon, UCRSuF = upper Columbia River summer–fall Chinook Salmon.

containing alumina-silica to remove highly polar compounds and were then further cleaned up using size-exclusion liquid chromatography. The sample extracts were then analyzed by low-resolution gas chromatography/mass spectrometry for PCB congeners, PBDE congeners, and organochlorine (OC) pesticides, including DDTs, hexachlorocyclohexanes (HCHs), chlordanes, aldrin, dieldrin, mirex, and endosulfans, as described by Sloan et al. (2005). Summed PCBs were determined by adding the concentrations of 45 congeners (PCBs 17, 18, 28, 31, 33, 44, 49, 52, 66, 70, 74, 82, 87, 95, 99, 101/90, 105, 110, 118, 128, 138/163/164, 149, 151, 153/132, 156, 158, 170/190, 171, 177, 180, 183, 187, 191, 194, 195, 199, 205, 206, 208, and 209). Additionally, concentrations of Cl3, Cl4, Cl5, Cl6, Cl7, Cl8, Cl9, and Cl10 CBs, as well as concentrations of dioxin-like PCBs (CBs 66, 74, 105, 118, and 156) were calculated to examine patterns of PCB homologs and proportions of dioxin-like PCBs in diets and fish samples from different sites, stocks, and life history types. Summed DDT levels ( $\sum$ DDTs) were calculated by summing the concentrations of p,p'-DDT, p,p'-DDE, p,p'-DDD, o, p'-DDD, o, p'-DDE, and o, p'-DDT. Summed chlordanes  $(\sum CHLDs)$  were determined by adding the concentrations of heptachlor, heptachlor epoxide, g-chlordane, a-chlordane, oxychlordane, cis-nonachlor, trans-nonachlor, and nonachlor III. Summed hexachlorocyclohexanes ( $\Sigma$ HCHs) were calculated by adding the concentrations of  $\alpha$ -HCH,  $\beta$ -HCH,  $\gamma$ -HCH, and lindane. In calculating sums, values < LOQ were treated as 0.

#### **Statistical Methods**

Statistical analyses were conducted with the JMP statistical package (SAS Institute, Inc., Cary, North Carolina). Differences in tissue and stomach contents contaminant concentrations by stock, site, and life history type (i.e., subyearlings versus yearlings) were determined by analysis of variance (ANOVA) and the Tukey-Kramer multiple range test, which is the preferred method of means comparison when sample sizes are not equal across treatment groups (Zar 1999). A two-factor ANOVA was used to examine the influence of stock and region of collection (i.e., in or below the Columbia Gorge) on body contaminant concentrations. Sites within the Columbia Gorge included Warrendale, Pierce Island, Franz Lake, and Mirror Lake, while sites below the Columbia Gorge included Confluence Washington, Confluence Oregon, Campbell Slough, Sandy Island, Columbia City, Beaver Army Terminal, Lord/Walker Island, White Island, Ryan Island, and Point Adams. Differences in the genetic composition of fish collected from different sampling sites, and proportions of fish above toxic-effects thresholds, were determined using contingency tables and chi-square analysis (Zar 1999). Data were log transformed as necessary

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to achieve a normal distribution. The significance level for all analyses was set at  $\alpha = 0.05$ .

## RESULTS

#### **Genetic Stock Identification**

Chinook Salmon from several of Columbia River stocks were represented in our sampling of unmarked subvearling Chinook Salmon in the lower Columbia River and estuary for chemical analyses (Figure 3). Approximately 91% of individuals were assigned to one of the genetic groups in our analysis, and relative probabilities were greater than 0.90. Assignments of the remaining samples (9%) were split between genetically similar groups (e.g., West Cascade fall and Spring Group fall). Of the subyearling Chinook Salmon we sampled 67% were estimated to be from the lower Columbia River ESU, 70% of those belonging to the West Cascades tributary fall stock and the other 30% to the Spring Creek Group fall stock. An additional 24% of subyearlings belonged to the upper Columbia summer-fall stock group. Spring-run subyearlings from the Willamette River stock accounted for an additional 7%, and the Snake River fall and Deschutes summer-fall stocks accounted for the remaining 2%. Generally the sites closest to the mouth of the estuary had the highest proportion of fish from the lower Columbia River ESU, although stocks from other ESUs were also represented. Sites closer to the Columbia Gorge had higher proportions of fish from the upper Columbia summer–fall and Snake River fall stocks. Spring-run fish from the upper Willamette River stock were found primarily at the Morrison Street Bridge site, in the Willamette River near downtown Portland. Yearling Chinook salmon, collected from the mouth of the estuary, were from the Snake River spring and mid and upper Columbia River spring stocks.

## Whole Body Lipid Content

*Yearlings versus subyearlings.*—Lipid content was significantly lower in body samples from yearling Chinook Salmon than in body samples from subyearlings (one-way ANOVA, P = 0.0116; Table 4). Lipid content of yearling Chinook Salmon ranged from 0.36% to 1.2%, with a mean of 0.71% (SD, 0.31), while in subyearling Chinook Salmon, lipid content ranged from 0.55% to 5.4%, with a mean of 1.6% (SD, 0.9).

Variation among stocks.—Because of the small number of Deschutes River summer–fall and Snake River fall Chinook Salmon in our samples, fish from these two ESUs were grouped for analyses of variation in lipid content among stocks. This

TABLE 4. Mean lipid content (%) in whole bodies (with stomach contents removed) of juvenile Chinook Salmon collected from the lower Columbia River. Samples are grouped by site of collection, genetic stock, and life history type. Values with different lowercase letters are significantly different (ANOVA and Tukey's multiple range test, P < 0.05). Stock abbreviations: UCR SuF = upper Columbia Summer–Fall, LCR-WCF = lower Columbia River–West Cascades Fall, LCR-SCGF = lower Columbia River Spring Creek Group, WR Sp = Willamette River Spring, UCR Sp = upper Columbia River Spring, Snake Sp = Snake River Spring.

Site	Percent lipid (±SD)	Stock	Percent lipid (±SD)		
Subyearlings					
		Snake Fall $(n = 4)$	$2.0~\pm~0.2~\mathrm{z}$		
Warrendale $(n = 5)$	$2.5~\pm~0.3$ zy	UCR SuF $(n = 20)$	$1.5~\pm~0.7~z$		
Pierce Island $(n = 1)$	1.1 zyx	LCR-WCF ( $n = 37$ )	$1.6 \pm 1.0$		
Franz Lake $(n = 2)$	$0.9~\pm~0.5~\mathrm{zyx}$	LCR-SCGF ( $n = 21$ )	$1.5~\pm~0.8~z$		
Mirror Lake $(n = 5)$	$1.2 \pm 0.3 \text{ zyx}$	WRSp $(n = 6)$	$1.6~\pm~0.5~z$		
Morrison St. Bridge $(n = 4)$	$1.7 \pm 0.4 \text{ zyx}$	UCRSp $(n = 2)$	$0.8~\pm~0.8~{ m zy}$		
Confluence WA ( $n = 12$ )	$1.4 \pm 0.2 \text{ yx}$	Snake Sp $(n = 5)$	$0.7~\pm~0.2~{ m y}$		
Confluence OR $(n = 7)$	$1.3 \pm 0.3 \text{ yx}$				
Campbell Slough $(n = 9)$	$1.6 \pm 0.3 \text{ zyx}$				
Sandy Island $(n = 6)$	$1.0 \pm 0.3 \text{ zyx}$				
Columbia City $(n = 7)$	$1.9 \pm 0.3 \text{ yx}$				
Beaver Army Terminal $(n = 10)$	$1.4 \pm 0.2 \text{ yx}$				
Lord/Walker Island $(n = 2)$	$1.1~\pm~0.2~\mathrm{zyx}$				
White Island $(n = 3)$	$1.5 \pm 0.1 \text{ zyx}$				
Ryan Island ( $n = 6$ )	$1.1 \pm 0.4 \text{ yx}$				
Point Adams $(n = 9)$	$2.7 \pm 0.3 z$				
Yearlings					
Trestle Bay $(n = 3)$	$0.6 \pm 0.4 \text{ yx}$				
McGowan $(n = 4)$	$0.8 \pm 0.4 \text{ x}$				
Yearlings from all sites $(n = 7)^a$			$0.7 \pm 0.3$		
Subyearlings from all sites $(n = 88)^a$			$1.6 \pm 0.9$		

<sup>a</sup>Yearlings and subyearlings differed significantly

grouping is consistent with the considerable genetic, life history, and ecological similarities of these two stocks, which were included in the same ESU in an early ESA status review (Myers et al. 1998). Among samples of subyearlings from fall Chinook Salmon stocks and Willamette River spring Chinook Salmon, mean lipid content ranged from 1.5% to 2.0%, while mean lipid content in the yearling Chinook Salmon from the mid and upper Columbia River spring and Snake River spring stocks was somewhat lower (0.67–0.81%; Table 4). Lipid content did not differ significantly among subyearlings from the fall Chinook Salmon stocks, but lipid content of the Snake River spring yearlings was significantly lower than lipid content of the summer and fall subyearlings from the Snake River, upper Columbia, West Cascades, and Spring Creek Group stocks ( $P \le 0.05$ ; Table 4).

Variation among sites.—Mean lipid content in whole body composites of subyearling Chinook Salmon from the lower Columbia River and estuary sampling sites ranged from <1% at Franz Lake and Sandy Island to 2.5–2.6% at Point Adams and Warrendale (Table 4). Values at these two sites were significantly different from each other, while other sites were intermediate (ANOVA and Tukey's LSD test, P < 0.05). Lipid levels in subyearling Chinook Salmon from Confluence-Oregon, Confluence-Washington, Sandy Island, and Ryan Island were significantly lower than lipid levels in subyearlings from Point Adams. Lowest lipid levels were found in yearlings from the McGowan and Trestle Bay sites. Of all Chinook Salmon sampled, the yearlings from Trestle Bay had the lowest lipid content (mean = 0.55%, SD = 0.43).

#### **Contaminant Concentrations in Salmon and Salmon Diets**

Chemical contaminants in salmon stomach contents.— Because of permit limitations on the number of salmon that could be collected in our study, a sufficient mass of stomach contents for chemical analysis could not be collected from all sites. Measurable concentrations of DDTs and PCBs were found in stomach contents of fish from all sampling sites where sufficient material could be collected (i.e., Warrendale, Morrison Street Bridge, Confluence-Washington, Campbell Slough, Columbia City, Sandy Island, Beaver Army Terminal, Point Adams, and West Sand Island; Figure 4). Concentrations of  $\Sigma$ PCBs were highest in stomach contents of fish collected from Morrison Street Bridge and Confluence (about 100 ng/g wet weight [wwt]), and lowest in stomach contents of salmon from West Sand Island and Warrendale (2-13 ng/g wwt). Concentrations of  $\sum PCBs$  (Figure 4a) were also generally low in fish stomach contents from Beaver Army Terminal (<13 ng/g wwt), except for of a single composite in which concentrations of  $\sum$ PCBs were 410 ng/g wwt. Concentrations of  $\sum$ PCBs in stomach contents of fish from other sites were fairly similar, ranging from 28 to 37 ng/g wwt. The PCB homologue profiles were very similar in samples from Morrison Street Bridge, Columbia City, Sandy Island, Beaver Army Terminal, and Point Adams (data not shown). The C15 and C16 PCBs were most abundant, together making up 65–85% of  $\sum$  PCBs at most of these sites and 100% of  $\sum$ PCBs at West Sand Island. In fish from Confluence-Washington and Campbell Slough, however, the PCB profile was different, stomach contents samples containing a higher proportion of C14 PCBs than those from the other sites. The proportions of dioxin-like PCBs in samples (CBs 66, 74, 77, 105, 118, 123, 156) ranged from 12% to 27%, higher percentages (23–27%) found at Campbell Slough, Confluence-Washington, and Beaver Army Terminal.

Concentrations of DDTs (Figure 4b) were highest in stomach contents of fish from Point Adams, but comparable concentrations of DDTs were also found in stomach contents of fish from Warrendale, the Confluence, and Columbia City. Concentrations of DDTs were lowest in stomach contents of Chinook Salmon from West Sand Island and Morrison Street Bridge. At all sites the predominant DDT isomers were p, p'-DDD and p, p'-DDE. In most of the samples analyzed, p,p'-DDE accounted for 70–80% of  $\sum$ DDTs, and *p*,*p*'-DDD accounted for 15–25% of  $\sum$ DDTs. Other isomers typically accounted for 1–3% of  $\sum$ DDTs. Stomach contents from Columbia City and Morrison Street Bridge deviated somewhat from this pattern; at Columbia City o,p'-DDD made up 12% of  $\sum$ DDTs, and p,p-DDE and p,p'-DDE only 54% of  $\sum$ DDTs; at Morrison Street Bridge, *p*,*p*'-DDD accounted for only 6% of  $\sum$ DDTs and *p*,*p*'-DDT accounted for 18%.

In addition to DDTs and PCBs, chlordanes, HCB and HCHs were detected in stomach contents of Chinook Salmon collected from several sites, highest concentrations being generally present at the Morrison Street Bridge, Columbia City, and Point Adams (Table 5). Chlordanes and HCHs were present at mean concentrations up to 5–8 ng/g wwt, while concentrations of HCBs were lower, typically <1 ng/g wwt. Other pesticides (e.g., aldrin, dieldrin, mirex, endosulfans) were below the detection limits (<0.1 ng/g wwt) in stomach contents of fish from all sites.

#### Persistent Organic Pollutants in Chinook Salmon Bodies

Variation by life history type.—Mean concentrations of DDTs were significantly higher (one-way ANOVA, P 0.024) in yearling (3,800 ng/g lipid, SD = 1,800, N = 7) than in subyearling (2,200 ng/g lipid, SD = 1,500, N = 88) Chinook Salmon. In both subyearlings and yearlings, the predominant DDT isomers in bodies were p,p'-DDD, which accounted for 14–16% of  $\sum$ DDTs, and *p*,*p*'-DDE, accounting for 76–81% of  $\sum$ DDTs. Other isomers typically accounted for <1-3% of  $\Sigma$ DDTs. In contrast to  $\Sigma$ DDTs, mean ( $\pm$ SD) lipid-adjusted  $\sum$ PCB concentrations were significantly lower (one-way ANOVA, P = 0.0074) in yearlings (4,100 ng/g lipid, SD = 1,000, N = 7) than subyearlings [1,300 ng/g lipid, SD =690, N = 88]. The PCB homologue profiles of yearlings and subyearlings were also somewhat different. In yearlings, the C15 and C16 PCBs were most abundant, together making up 79% of  $\sum$  PCBs. Subyearlings had higher proportions of Cl3 and Cl4 PCBs, this group making up 45% and the Cl5 and Cl6 PCBs making up 49% of  $\sum$  PCBs. The proportion of



FIGURE 4. Mean (+SD) concentrations of (a)  $\Sigma$ PCBs (polychlorinated biphenyls) and (b)  $\Sigma$ DDTs (dichlorodiphenyltrichloroethane) in stomach contents of subyearling Chinook Salmon samples of various sites in the lower Columbia River and estuary. Sites are ordered from upstream to downstream, the Warrendale site being the farthest from the mouth of the Columbia. Values with different lowercase letters are significantly different (one-factor ANOVA and Tukey's LSD test; *P* < 0.05).

TABLE 5. Mean concentrations of organochlorine pesticides in whole bodies and stomach contents of juvenile Chinook Salmon collected from the lower Columbia River. Values with different lowercase letters are significantly different (ANOVA and Tukey's multiple range test, P < 0.05). Abbreviations: < LOQ (limits of quantitation) = below limits of quantitation (0.16 – 0.39 ng/g wet weight),  $\sum$ HCH = summed hexachlorocyclohexanes (includes  $\alpha$ -HCH,  $\beta$ -HCH, and  $\gamma$ -HCH [lindane]),  $\sum$ CHLDs = summed chlordanes (includes heptachlor, heptachlor epoxide,  $\gamma$ -chlordane, oxychlordane, *cis*-nonachlor, trans-nonachlor and nonachlor III); HCB = hexachlorobenzene. In addition, Mirex, endosulfans, and aldrin were measured but were below detection limits in all samples.

Site	$\sum$ HCH ± SD	Dieldrin $\pm$ SD	$\sum$ CHLDs $\pm$ SD	HCB $\pm$ SD		
Salmon bodies – subyearlings (ng/g lipid)						
Warrendale $(n = 5)$	< LOQ z	< LOQ y	$21 \pm 21 \text{ y}$	$16 \pm 7 \text{ yx}$		
Pierce Island $(n = 1)$	< LOQ z	< LOQ zy	< LOQ zy	33 zyx		
Franz Lake $(n = 2)$	< LOQ z	< LOQ y	$18 \pm 25 \text{ zy}$	$11 \pm 15 \text{ yx}$		
Mirror Lake $(n = 5)$	< LOQ z	< LOQ y	$31 \pm 13 \text{ y}$	$10 \pm 11 \text{ x}$		
Morrison St. Bridge $(n = 4)$	< LOQ z	$13 \pm 26 \text{ y}$	$230 \pm 150 \text{ zy}$	$93 \pm 42 z$		
Confluence, Washington $(n = 12)$	< LOQ z	4 ± 7 y	$93 \pm 62 \text{ y}$	$28 \pm 14 \text{ yx}$		
Confluence, Oregon $(n = 7)$	< LOQ z	$42 \pm 53 \text{ zy}$	$280 \pm 300 z$	$43 \pm 27 \text{ yx}$		
Campbell Slough $(n = 9)$	< LOQ z	$16 \pm 18 \text{ y}$	$26 \pm 11 \text{ y}$	$18 \pm 9 \mathrm{x}$		
Sandy Island $(n = 6)$	< LOQ z	$110 \pm 82 z$	$32 \pm 10 \text{ y}$	$27 \pm 6 \text{ yx}$		
Columbia City $(n = 7)$	< LOQ z	$20 \pm 19 \text{ y}$	$150 \pm 110 \text{ zy}$	$25 \pm 13 \text{ yx}$		
Beaver Army Terminal $(n = 10)$	< LOQ z	$20~\pm~24~{ m y}$	$150 \pm 77 \text{ zy}$	$28 \pm 9 \text{ yx}$		
Lord/Walker Island $(n = 2)$	< LOQ z	< LOQ y	$230 \pm 50 \text{ zy}$	$50 \pm 15 \text{ zyx}$		
White Island $(n = 3)$	< LOQ z	$14 \pm 4 y$	$68 \pm 25 \text{ zy}$	$32 \pm 3 \text{ yx}$		
Ryan Island ( $n = 6$ )	< LOQ z	< LOQ y	$58~\pm~28~{ m y}$	$31 \pm 9 \text{ yx}$		
Point Adams $(n = 9)$	< LOQ z	$8 \pm 8 \text{ y}$	93 ± 37 y	$23 \pm 4 \text{ yx}$		
Salmon bodies – yearlings (ng/g wet weight)						
Trestle Bay $(n = 3)$	< LOQ y	< LOQ y	$140 \pm 51 \text{ zy}$	$40 \pm 37 \text{ yx}$		
McGowan $(n = 4)$	< LOQ y	$18~\pm~22~{ m y}$	$140 \pm 60 \text{ zy}$	$54~\pm~19~zy$		
Stomach contents						
Warrendale $(n = 5)$	< LOQ y	< LOQ	$0.4~\pm~0.6~{ m zy}$	$0.2 \pm 0.3 \text{ yx}$		
Morrison Street Bridge $(n = 7)$	$1.6 \pm 3.0 \text{ zy}$	< LOQ	$1.1 \pm 1.2 \text{ zy}$	$0.7~\pm~0.5$ zy		
Confluence WA $(n = 5)$	< LOQ y	< LOQ	$1.4 \pm 3.1 \text{ zy}$	$0.1 \pm 0.2 \text{ yx}$		
Campbell Slough $(n = 3)$	< LOQ zy	< LOQ	$0.1 \pm 0.3 \text{ zy}$	< LOQ yx		
Columbia City $(n = 6)$	$7.4~\pm~8.8~z$	< LOQ	$4.1\pm~5.4$ zy	$0.5~\pm~0.5~{ m zyx}$		
Sandy Island $(n = 3)$	< LOQ zy	< LOQ	$1.3 \pm 1.5 \text{ zy}$	$0.4 \pm 0.4 \text{ zyx}$		
Beaver Army Terminal $(n = 6)$	$0.2 \pm 0.5$	< LOQ	$0.1~\pm~0.3~{ m y}$	$0.1 \pm 0.2 \text{ x}$		
Point Adams $(n = 10)$	$0.3~\pm~0.6~\mathrm{y}$	< LOQ	$5.1 \pm 2.4 z$	$0.9\pm0.4$ z		
West Sand Island $(n = 1)$	< LOQ zy	< LOQ	< LOQ zy	< LOQ zyx		

dioxin-like PCB congeners (CBs 66, 74, 177, 105, 118, 114, 123, 156, 157, 167, and 189) was about the same in yearlings (15%) and subyearlings (16%).

Variation among stocks.—Among stocks,  $\sum$ PCB concentrations tended to be highest in Spring Creek Group fall, upper Columbia summer-fall, and Snake River fall Chinook Salmon, and lowest in Snake River spring and mid and upper Columbia River spring Chinook Salmon (Figure 5a). However, concentrations measured in all stocks were extremely variable, and there were no significant differences among the stocks (ANOVA, P = 0.2726). The PCB homologue profiles of the whole bodies were similar mid and upper Columbia spring, Snake River spring, and West Cascades fall Chinook Salmon. The C15 and C16 PCBs were most abundant, together making up 75–85% of total PCBs. Snake River fall Chinook Salmon were comparable but

had somewhat higher proportions of Cl3 and Cl4 PCBs (38%) and lower proportions of Cl5 and Cl6 PCBs (56%). However, the PCB profiles were quite different in Spring Creek Group fall Chinook Salmon, with fish from these stocks containing a much higher proportion of Cl3 and Cl4 PCBs ( $59\%\sum$ PCBs) than fish from the other sites. The proportion of dioxin-like PCB congeners (CBs 66, 74, 177, 105, 118, 114, 123, 156, 157, 167, and 189) was about the similar in all stocks, ranging from 13% in mid and upper Columbia River spring Chinook Salmon to 17% in Snake River fall Chinook Salmon.

Concentrations of DDTs tended to be higher in Snake River spring and mid and upper Columbia spring Chinook Salmon and lower in Willamette River spring Chinook Salmon (Figure 5b). Again, however, differences were not statistically significant (ANOVA, P = 0.3872). In most of the samples analyzed of all



FIGURE 5. Mean (+SD) concentrations of (a)  $\Sigma$ DDTs (polychlorinated biphenyls) and (b)  $\Sigma$ PCBs (dichlorodiphenyltrichloroethane) in Chinook Salmon samples of different stocks from the lower Columbia River and estuary. Subyearlings are represented in black and yearlings are represented in white. No significant differences were observed among stocks for  $\Sigma$ DDTs or  $\Sigma$ PCBs (one-factor ANOVA and Tukey's LSD test; *P* < 0.05). See Figure 3 for population abbreviations.

Chinook Salmon stocks, the predominant DDT isomers in bodies were p,p'-DDD, which accounted for 13–17% of  $\sum$ DDTs, and p,p'-DDE, which accounted for 75–85% of  $\sum$ DDTs. Other isomers typically accounted for 1–5% of  $\sum$ DDTs.

Variation among sites.—Concentrations of  $\sum$ PCBs were lowest in subyearling Chinook Salmon from sites in the Columbia Gorge, where mean  $\sum$ PCB concentrations ranged from 360 to 930 ng/g lipid (Figure 6a). These levels were slightly lower than concentrations measured in yearling spring Chinook Salmon from the Trestle Bay and McGowan sites near the mouth of the estuary (1,100–1,500 ng/g lipid wwt). Concentrations of PCBs were highest at the Confluence-Oregon, Confluence-Washington, and Beaver Army Terminal sites: range, 5,400–12,000 ng/g lipid. In Chinook Salmon from the Morrison Street Bridge site in the lower Willamette River near downtown Portland, Oregon, mean  $\sum$ PCB concentrations were 1,500 ng/g lipid. In subyearling Chinook Salmon from the other sites downstream of the Willamette–Columbia confluence (Campbell Slough, Sandy Island, Lord/Walker Island, Ryan Island, White Island, and Point Adams), concentrations of  $\sum$ PCBs ranged from 940 to 2300 ng/g lipid. The PCB homologue profiles of the whole bodies were similar in fish from



FIGURE 6. Mean (+SD) concentrations of (a)  $\Sigma$ PCBs (polychlorinated biphenyls) and (b)  $\Sigma$ DDTs (dichlorodiphenyltrichloroethane) in Chinook Salmon samples from different sites in the lower Columbia River and estuary. Subyearlings are represented in black and yearlings are represented in white. Values with different lowercase letters are significantly different (one-factor ANOVA and Tukey's LSD test; P < 0.05).

Warrendale, Pierce Island, Franz Lake, Mirror Lake, Morrison Street Bridge, Columbia City, Sandy Island, Beaver Army Terminal, Point Adams, McGowan, and Trestle Bay (data not shown). The C15 and C16 PCBs were most abundant, together making up 75–85% of total PCBs. Fish from the Confluence-

Washington, Campbell Slough, Ryan Island, and Whites Island sites were comparable but had somewhat higher proportions of Cl3 and Cl4 PCBs (27–32%) and lower proportions of Cl5 and Cl6 PCBs (55–65%). In fish from the Confluence-Oregon, however, the PCB profile was quite different, these fish containing

a higher proportion of Cl3 and Cl4 PCBs (80% of total PCBs) than fish from the other sites. The proportion of dioxin-like PCB congeners was lower in samples from sites in the Columbia Gorge (12%) than in samples from sites below the Gorge (16%). Samples from the Confluence-Washington and Campbell Slough sites had the highest proportions of dioxin-like PCBs (21–22%).

With the exception of the Mirror Lake site, concentrations of DDTs in subyearling Chinook Salmon tended to be lowest in at sites in the Columbia Gorge, where mean DDT concentrations ranged from 1,100 to 1,500 ng/g lipid (Figure 6b). Mean DDT concentrations were highest at Mirror Lake, Confluence Oregon, Confluence Washington, and Beaver Army Terminal, ranging from 3,000 to 3,200 ng/g lipid (Figure 6b). In Chinook Salmon from the Morrison Street Bridge site in the lower Willamette River near downtown Portland, mean DDT concentrations were 1,300 ng/g lipid. In subyearling Chinook Salmon from the sites downstream of the Willamette-Columbia confluence other than Beaver Army Terminal (i.e., Campbell Slough, Sandy Island, Lord/Walker Island, Ryan Island, White Island, and Point Adams) mean concentrations of DDTs ranged from 1,500 to 2,100 ng/g lipid. These levels were lower than mean DDT concentrations measured in yearling spring Chinook Salmon from the Trestle Bay and McGowan sites near the mouth of the estuary, which ranged from 3,700 to 4,100 ng/g lipid. At all sites the predominant DDT isomers in bodies were p,p'-DDD and p,p'-DDE. In most of the samples analyzed, p,p'-DDE accounted for 70–80% of  $\sum$ DDTs, and *p*,*p*'-DDD accounted for 15–25% of  $\sum$ DDTs. Other isomers typically accounted for 1–5% of  $\sum$ DDTs. The proportion of p,p'-DDT was somewhat higher in fish from the Morrison Street Bridge site (8% of  $\sum$ DDTs) than in fish from the other sites.

In addition to PCBs and DDTs, chlordanes, hexachlorobenzene, and dieldrin were also detected in whole bodies of estuarine Chinook Salmon from one or more sampling sites, but at much lower concentrations than PCBs or DDTs (Table 5). Of the pesticides detected, chlordanes were generally found at the highest concentrations, mean concentrations ranging from 18 to 280 ng/g lipid. Other organochlorine pesticides (i.e., hexachlorocyclohexanes, mirex, aldrin, and endosulfans) were below the limits of quantitation.

Columbia Gorge versus below the Gorge.—Because of the relatively high concentrations of  $\sum$ PCBs and DDTs measured in Chinook Salmon sampled from sites around and below Portland and Vancouver in comparison with Chinook Salmon sampled from the Columbia Gorge sites, concentrations of these contaminants were compared in Chinook Salmon from various stocks collected in the Gorge and below the Gorge after they had passed through and been exposed to contaminants in the Portland–Vancouver urban and industrial area.

For  $\sum$  PCBs, two-factor ANOVA indicated that capture in versus below the Gorge had a significant influence on lipid-adjusted  $\sum$  PCB concentrations (P = 0.0178), but stock of origin did not (P = 0.2168; Figure 7a). The mean  $\sum$  PCB con-



FIGURE 7. Mean (+SD) concentrations of (a)  $\Sigma$ PCBs (polychlorinated biphenyls) and (b)  $\Sigma$ DDTs (dichlorodiphenyltrichloroethane) in Chinook Salmon samples from different stocks in the lower Columbia River and estuary collected in the Columbia Gorge and below the Columbia Gorge. Two-way analysis of variance indicated capture in versus below the Columbia Gorge had a significant effect on  $\Sigma$ PCB concentrations (0.0001 < *P* < 0.0178). Neither area of capture nor stock of origin significantly affected  $\Sigma$ DDT concentrations (0.1014 < *P* < 0.7182). Values in parentheses separated with a comma represent the number of samples above and below, respectively, the Columbia Gorge for each stock. See Figure 3 for abbreviations.

centration in Chinook Salmon from the Gorge sites was 740 ng/g lipid wwt versus 4,600 ng/g lipid wwt for sites below the Gorge. For all stocks for which subyearlings were captured both above and below the Gorge (Spring Creek Group fall, Snake River fall, and upper Columbia River summer/fall Chinook Salmon) concentrations of  $\sum$ PCBs were much higher in fish below than above the Gorge (Figure 7a). Subyearling Chinook Salmon collected from the Gorge sites also had lower proportions of Cl3 and Cl4 PCBs (16% of  $\sum$ PCBs) than did subyearling Chinook Salmon collected below the Gorge (46% of  $\sum$ PCBs).

For DDTs, two-factor ANOVA indicated that neither capture in versus below the Gorge (P = 0.1014) nor stock of origin (P = 0.7182) had a significant influence on DDT concentrations (Figure 7b). The mean DDT concentration in Chinook Salmon from the Gorge sites was 2,100 ng/g lipid versus 2,200 ng/g lipid for sites below the Gorge. For Snake River fall and upper Columbia fall Chinook Salmon, concentrations of DDTs tended to be higher in fish below than above the Gorge, but for Spring Creek fall Chinook Salmon, DDTs were slightly higher in fish above than below the Gorge (Figure 7b). Distribution of DDTs within body samples was very similar in subyearlings sampled above and below the Gorge, p.p'-DDE accounting for 78–83% of  $\sum$ DDTs and p.p'-DDD accounting for 15–17%.

Among samples from different stocks collected below the Gorge, DDT concentrations showed considerable variability, mean values ranging from 1,300 ng/g lipid in Willamette River

spring Chinook Salmon to 5,300 ng/g lipid in Snake River fall Chinook Salmon. Concentrations of DDTs were similar in the two lower Columbia River stocks represented; for West Cascades fall Chinook Salmon the mean DDT concentration was 2,200 ng/g lipid; for Spring Creek Group fall Chinook Salmon it was 2,000 ng/g lipid. However,  $\sum$ DDTs,  $\sum$ PCB concentrations among stocks were not significantly different (P = 0.1015).

Among samples from different stocks collected below the Gorge,  $\sum$ PCB concentrations were even more variable than concentrations of DDTs. Mean values on a lipid-adjusted basis ranged from 1,800 ng/g lipid in Willamette River spring Chinook Salmon to 11,000 ng/g lipid in Snake River fall Chinook Salmon. In contrast to DDTs, concentrations of  $\sum$ PCBs were substantially higher in Spring Creek Group fall Chinook Salmon (6,900 ng/g lipid wwt) than in West Cascades fall Chinook Salmon (3,100 ng/g lipid). However, as with DDTs,  $\sum$ PCB concentrations among stocks were not significantly different (P = 0.0891).

## DISCUSSION

We measured persistent organic pollutants in whole body and stomach content samples of juvenile Chinook Salmon from the Columbia River basin, PCBs and DDTs being the most abundant with regard to concentrations and frequency of detection. Chlorinated pesticides, including chlordanes, hexachlorobenzene, and dieldrin, were also measured in these samples but were found in lower concentrations and at lower frequencies than PCBs and DDTs. Concentrations of PCBs in salmon body samples ranged from 22 to 69,00 ng/g lipid, while concentrations of DDTs ranged from 78 to 7.500 ng/g lipid. In diet samples, concentrations of PCBs ranged from below detection limits to 410 ng/g wwt, whereas DDT levels ranged from 2.9 to 78 ng/g wwt. The PCB and DDT concentrations reported in the current study are generally consistent with those reported by Johnson et al. (2007a) in juvenile Chinook Salmon from the lower Columbia River and estuary, with the exception of some very high concentrations of DDTs (30,000-72,000 ng/g lipid) found in Chinook Salmon smolts collected near the estuary mouth in the earlier study. These high DDT concentrations are related, at least in part, to the very low lipid content of these fish (0.39–0.44%; Johnson et al. 2007a).

The lower concentrations of both DDTs and PCBs measured in this study are similar to levels measured in out-migrant juvenile Chinook Salmon body and diet samples from other nonurban estuaries in Oregon and Washington (Johnson et al. 2007b). Concentrations of PCBs in the higher range are comparable to those measured in juvenile salmon body and diet samples from urban and industrialized areas of Puget Sound (Stein et al. 1995; Stehr et al. 2000; Johnson et al. 2007b, Meador et al. 2010; Kelley et al. 2011). For example, concentrations of PCBs in juvenile Chinook Salmon bodies from Elliott Bay (Seattle, Washington) and Commencement Bay (Tacoma, Washington) ranged from 640 to 72,500 ng/g lipid while in diet samples from these same areas, concentrations of PCBs ranged from 45 to 445 ng/g wwt

(Stehr et al. 2000; Olson et al. 2008; Johnson et al. 2007b; Meador et al. 2010). Concentrations of DDTs in the juvenile Chinook Salmon sampled in this study were somewhat higher than concentrations measured in juvenile Chinook Salmon from other Pacific Northwest estuaries including Puget Sound (Stein et al. 1995; Stehr et al. 2000; Johnson et al. 2007b). In juvenile Chinook Salmon bodies from Elliott Bay and Commencement Bay, concentrations of DDTs in juvenile Chinook Salmon bodies ranged from 190 to 4,000 ng/g lipid and from 11 to 43 ng/g wwt in stomach contents samples (Stehr et al. 2000; Johnson et al. 2007b; Olson et al. 2008; Meador et al. 2010). While it would be interesting to compare these values with contaminant concentrations in juvenile salmon from river systems outside the Pacific Northwest, these data are lacking. A number of studies have been conducted on concentrations of bioaccumulative contaminants in adult salmon in other river systems (e.g., Jackson et al. 2001; Missildine et al. 2005; Kelly et al. 2007, 2011; Montory et al. 2010), but there is little comparable information on out-migrating juveniles.

Some perspective on the toxicity of the PCB and DDT concentrations measured in juvenile salmon bodies is provided by critical body residue values in two other studies. Meador et al. (2002) estimated an adverse health effects threshold for  $\sum PCBs$ of 2,400 ng  $\sum$  PCBs/g lipid, based on a wide range of toxicological studies on juvenile trout and salmon with effects ranging from enzyme induction to mortality. A similar threshold for  $\sum$ DDTs has not been developed that is specific to juvenile salmon. However, Beckvar et al. (2005) estimated that concentrations above 600 ng/g wwt or 6,000 ng/g lipid (adjusted for lipid content as recommended by Johnson et al. 2007b) may cause adverse effects in a variety of fish species, including Pacific salmon, based on literature values for end-points including survival, growth, and reproduction. This guideline may not be fully protective for DDTs because it includes limited data on sublethal endpoints, such as endocrine disrupting, immunotoxic, and behavioral effects (Beckvar et al. 2005), but provides a good starting point for evaluation. Of the salmon body samples we analyzed, only two (3.2%) were above the 6,000 ng/g lipid guideline for DDTs. On the other hand, 20 of the 62 samples (32%) had PCB concentrations above the 2,400 ng/g lipid guideline.

Because of the small size of juvenile salmon we were forced to perform analyses on composite samples. This reduces the sample size and affects sample variability, thus limiting the statistical rigor of our analyses. Nonetheless, we were able to observe some distinct patterns in contaminant concentrations among sites, stocks, and life history types for both PCBs and DDTs. As predicted, subyearlings overall had significantly higher concentrations of PCBs than yearlings. Concentrations of PCBs were at or above the 2,400 ng/g lipid critical body residue estimated by Meador et al. (2002) in 33% of subyearling Chinook Salmon samples versus 14% of yearlings analyzed as part of our study. Also as expected, spring Chinook Salmon yearlings had higher concentrations of DDTs than subyearlings, 14% of samples exceeding the threshold of 6,000 ng/g lipid (Beckvar et al. 2005; Johnson et al. 2007b) versus 1.1% of samples from subyearlings. These findings are consistent with our hypothesis that urban-associated contaminants, represented by PCBs, would be present at higher concentrations in subyearling migrants, while yearling migrants originating primarily in the interior Columbia and Snake river basins would be more likely to accumulate agricultural contaminants, represented by DDTs.

We also observed lower lipid content in yearlings (0.36– 1.3%) than in subyearlings (0.55-5.4%). The extended migration from headwater rearing areas and stress of smoltification typically lead to loss of body lipids in juvenile salmon (Sheridan 1989), so it is not surprising that this would be apparent in the spring Chinook Salmon captured in the estuary just before ocean entry. In our earlier study (Johnson et al. 2007a), we also noted low body lipid content in subyearling and yearling smolts sampled from the mouth of the Columbia River. Similarly, Arkoosh et al. (2011) observed significant lipid loss in spring Chinook Salmon during out-migration from the Snake River to Bonneville Dam and further noted that the reduction in lipid content led to a corresponding increase in lipid-adjusted body concentrations of PCBs and DDTs, even in cases where changes in wet weight concentrations were minimal. In our study as well, very low body lipid content contributed to relatively high lipidadjusted PCB and DDT concentrations in some samples. Similar relationships between lipids and bioaccumulative contaminants have been observed in returning adult Pacific salmon (deBruyn et al. 2004; Kelly et al. 2007). Body lipid content can influence an organism's tolerance of bioaccumulative contaminants, individuals with lower lipid content typically showing a greater toxic response to comparable exposure (Lassiter and Hallam 1990). Consequently, low lipid levels and declines in body lipid content, such as those observed in outmigrating juvenile salmon, may increase their susceptibility to the toxic effects of contaminants such as PCBs and DDTs.

In addition to differences between yearling and subyearlings, distinct patterns of contaminant accumulation were also observed in different Chinook Salmon stocks, particularly in the case of PCBs. Concentrations of PCBs measured in different Chinook Salmon stocks generally reflected their use of the lower Columbia River, particularly the region around Portland and Vancouver. The highest concentrations of PCBs were observed in subyearlings from the three fall Chinook Salmon stocks that migrate through the Portland-Vancouver area and use these locales for feeding and rearing: Snake River fall Chinook Salmon, upper Columbia summer-fall Chinook Salmon, and lower Columbia River's Spring Creek Group fall Chinook Salmon. From 33% to 50% of samples from these stocks had PCB concentrations above the 2,400-ng/g lipid threshold. Concentrations of PCBs were somewhat lower in subyearlings from the lower Columbia West Cascades fall Chinook Salmon stock, which includes many fish that originate in lower Columbia River watersheds below the Portland-Vancouver area, and 30% of samples from this stock had PCB concentrations exceeding 2,400 ng/g lipid.

Additional results point to the lower Willamette River and the Vancouver–Portland metropolitan areas as important sources of Chinook Salmon exposure to industrial contaminants, as represented by PCBs. Concentrations of PCBs were highest in salmon diet samples from sites in this area, and the elevated PCB concentrations in Chinook Salmon prey were generally reflected in PCB concentrations in Chinook Salmon collected from the Willamette–Columbia Confluence sites, on both the Washington and Oregon sides of the river, had the highest PCB concentrations observed (average, 11,000–12,000 ng/g lipid).

We also observed a tendency for fish collected at sites from the Portland–Vancouver area or directly downstream to have higher proportions of Cl3 and Cl4 PCBs than fish collected from other sites. This homolog pattern was also observed more commonly in subyearlings than yearlings and more in Willamette River spring and Spring Creek Group fall Chinook Salmon than in other stocks. All of these groups of fish are among those most likely to have spent time feeding and rearing in the urbanized portions of the lower Willamette and lower Columbia rivers.

The importance of the Portland-Vancouver area as a source of PCB contamination is further emphasized by the fact that, in fish from all stocks sampled in both regions, PCB concentrations were lower in samples collected in the Columbia Gorge, which is above the Willamette-Columbia Confluence and the Portland-Vancouver metropolitan area, than in samples collected in or below the Portland-Vancouver area. No Chinook Salmon sampled from above the Confluence had PCB concentrations exceeding the 2,400 ng/g lipid threshold, whereas 36% of samples collected at or below this region had PCB concentrations above the threshold. Subyearling Chinook Salmon collected from above the Confluence also had lower proportions of trichlorinated and tetra-chlorinated PCBs and lower proportions of dioxin-like PCBs (e.g., PCBs 118, 156) than those collected at or below this region. The finding that PCBs tend to be higher in juvenile Chinook Salmon from all stocks captured below the Portland–Vancouver area than in those captured in the Columbia Gorge was similar to the pattern observed by Sloan et al. (2010) for PBDEs, again highlighting the Portland-Vancouver area as a source of industrial and wastewater contaminants that appear to be impacting multiple stocks.

In contrast to PCBs, DDTs appeared to be distributed more uniformly throughout the lower Columbia River and estuary and were present in prey samples at concentrations of 20– 30 ng/g wwt at most sites. Similar to concentrations of  $\sum$ DDTs in stomach contents, concentrations of  $\sum$ DDTs in Chinook Salmon bodies were less variable based on site of collection than were PCBs, average concentrations ranging from 1,100 to 4,100 ng/g lipid. Sites where the highest concentrations were observed included some of the same sites where PCBs were elevated (Confluence-Oregon, Confluence-Washington, and Beaver Army Terminal), but concentrations of  $\sum$ DDTs were also relatively high in samples from some other areas, such as Mirror Lake in the Columbia Gorge. 38

As noted earlier, concentrations of DDTs were highest in the yearling migrants from spring Chinook Salmon stocks (upper Columbia River spring and Snake River spring), but among the other Chinook Salmon stocks examined, body concentrations of  $\sum$ DDTs showed no significant differences overall. Moreover, when body concentrations of  $\sum$ DDTs were compared in samples from various stocks collected in and below the Columbia Gorge, levels were generally similar in fish from both areas, suggesting that Chinook Salmon are absorbing DDTs before entering the lower Columbia River. This is consistent with multiple studies documenting DDT contamination in the interior Columbia basin (USEPA 2009) and with the findings of Arkoosh et al. (2011), both indicating uptake of DDTs in hatchery-origin Snake River spring Chinook Salmon migrating downriver through the Snake and middle Columbia rivers. The only exception to this trend were the Snake River fall Chinook Salmon subyearlings, in which concentrations of  $\sum$ DDTs were more than two times higher in samples collected below the Gorge than in those from within the Gorge. The fish in these particular samples may have been exposed to DDTs at multiple locations within and above the lower Columbia River. Indeed, some DDT accumulation in the lower river would be expected, considering the extent of agricultural land use in the Willamette basin, as well as the existence of a point source of DDT contamination within Portland Harbor (LWG 2007).

Surprisingly, neither PCBs nor DDTs were found at particularly high concentrations in Willamette River spring Chinook Salmon, in spite of extensive industrial, urban, and historical agricultural land uses in the Willamette basin. This may have been because the majority of our samples were collected from the Morrison Street Bridge site near downtown Portland at Willamette river kilometer (rkm) 22 (rkm 0 is at the confluence of the Willamette and Columbia rivers), upstream of the reach of the lower Willamette River encompassing the Portland Harbor Superfund site, where some of these highest concentrations of PCBs and DDTs have been found. Remedial investigation studies at the Portland Harbor Superfund site (LWG 2007) also found relatively low concentrations of PCBs and DDTs in juvenile Chinook Salmon upstream of the Superfund site, at Willamette rkm 29. In these fish, mean concentrations of PCBs and DDTs were 870 ng/g lipid and 490 ng/g lipid. In contrast, in juvenile Chinook Salmon sampled from sites within the Superfund area (Willamette rkm 11-16), concentrations of PCBs were as high as 11,300 ng/g lipid (rkm 16), while DDT concentrations were as high as 14,800 ng/g lipid (rkm 11). Although we found that PCB concentrations were elevated in stomach contents of fish from the Morrison Street Bridge site, the levels were also quite variable, so samples with high concentrations may not reflect the typical diet of fish at this site. Moreover, the likely residence time of juvenile Chinook Salmon at this site is not known. All of the samples we analyzed were collected from Willamette River spring subyearlings, and genetic analysis shows that Willamette River subyearling spring Chinook Salmon have a very protracted downstream dispersal and enter the lower river throughout the spring and early summer, as well as in the autumn (D.J.T., unpublished data). While some fish may spend several months feeding and rearing in the lower Willamette River and Columbia–Willamette confluence area (Friesen et al. 2005, Teel et al. 2009), residence times can be highly variable. Additional sampling of both subyearling and yearling Willamette River Spring Chinook Salmon from sites within the upper and lower Willamette River and lower Columbia River is needed to better characterize exposure patterns.

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