



Washington
Department of
**FISH and
WILDLIFE**

An Evaluation of the Efficacy of Remedial Actions Implemented in the Commencement Bay Nearshore and Tideflats Superfund Site to Reduce PCB Contamination: 1984-2019

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James E. West
Washington State Department of Fish and Wildlife
PO Box 43200
Olympia, WA 98504-3200
james.west@dfw.wa.gov
360.870.8303
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Executive Summary

The Washington Department of Fish and Wildlife (WDFW) conducted a field study in 2019 in the Commencement Bay Nearshore and Tideflats (CB/NT) Superfund site to evaluate whether polychlorinated biphenyls (PCBs) in English sole (*Parophrys vetulus*), a bottomfish, have declined sufficiently since 1984 to achieve EPA's target tissue cleanup objective for human health. This was determined by comparing PCBs in English sole sampled from 15 locations in Operable Unit 1 of the CB/NT Superfund site with English sole from Carr Inlet, a nearby reference location in the southern Puget Sound basin containing no known or suspected local PCB sources.

Two analytical methods for measuring PCBs and calculating their totals provided largely congruent results. Total PCBs measured using Aroclor-methods were required by the Record of Decision, however paired PCB-congener analysis methods on sample splits provided more clarity and certainty regarding the distribution of PCBs throughout the CB/NT. Congener-PCBs also allowed the opportunity to compare CB/NT English sole PCB results with other areas and species in Puget Sound. Results for both methods are compared and reported herein.

Although the overall CB/NT recovery goal was met (the average site-wide PCB concentration from all English sole in the 15 sampling locations of Operable Unit 1 was statistically indistinguishable from the Carr Inlet Reference Area (CIRA)), substantial areas of contamination in the CB/NT remained. English sole from two of the largest CB/NT waterways, Hylebos and Thea Foss, exhibited significantly greater PCB tissue concentrations than the CIRA, at concentrations among the highest PCB levels measured in English sole on a Puget Sound-wide scale (based on a comparison with English sole from WDFW's long-term Toxics Biological Observation System, or TBIOS). Moreover, long-term TBIOS monitoring of PCB levels in English sole from the Thea Foss Waterway has shown no evidence of a declining PCB trend over the past 30 years.

The status of Blair Waterway is less clear. Although the average PCB concentration in English sole from Blair was statistically indistinguishable from the CIRA fish, sole from the head of the Blair Waterway exhibited one of the two highest PCB concentrations in the entire study, and 44% of the samples taken from the head of the Blair were above the average CIRA concentration. These observations suggest English sole continue to accumulate PCBs in the Blair Waterway, particularly from its head, at concentrations that can greatly exceed the recovery goal. More sampling in the Blair Waterway may be necessary to solidify its status.

Continued contamination of the CB/NT area, especially Hylebos and Thea Foss Waterways has also been reported in other fish species reported by WDFW's TBIOS. Relatively high PCB concentrations in fish tissues have been observed in the Hylebos Waterway over six years from 2013 to the current study (2019) in juvenile Chinook salmon (*Oncorhynchus tshawytscha*), cutthroat trout (*O. clarkii clarkii*), and transplanted bay mussels (*Mytilus trossulus*). PCBs in cutthroat trout from the Hylebos exhibited the highest concentration of PCBs in any whole-body fish sampled by TBIOS over the past 30 years throughout Puget Sound, and the area surrounding the CB/NT industrial waterways has been reported as one of the most PCB-contaminated nearshore habitats for outmigrating juvenile Chinook salmon among twelve major Puget Sound rivers in recent years. Some PCB concentrations in both juvenile Chinook salmon and cutthroat trout from CB/NT locations were at levels reportedly high enough to impair their health or result in their mortality.

Continued contamination in fish from the Hylebos, Thea Foss and Blair Waterways is significant not only because of the risk to humans from consuming seafood from the CB/NT, but also because these waterways occupy a large area of productive nearshore and shoreline habitats in the CB/NT used by

many important species including juvenile salmon. Considering the four waterways that have not been filled-in¹, only Sitcum Waterway, the smallest in area of the four, had English sole which unambiguously met the human health recovery objective. Chinook salmon juveniles, a species listed as threatened under the US federal Endangered Species Act, regularly migrate through and feed in these habitats during their normal seaward migration from Puget Sound rivers (most likely the Puyallup and Rivers for salmon sampled from the CB/NT site), highlighting concern for contamination in the CB/NT beyond the human health objective.

PCB concentrations in fish have declined sufficiently in Sitcum Waterway, the nearshore area fronting the mostly in-filled St. Paul, Milwaukee, and Middle Waterway, and the three areas along the western shoreline of Commencement Bay (Old Town, Ruston, and Pt. Defiance) to meet the CB/NT human health recovery objective. One fish from the Pt. Defiance location exhibited a relatively high total Aroclor concentration (67 ng/g wet wt). However, its total PCB concentration measured using the PCB congener method was less than half that, at 30 ng/g wet weight, so the PCB result from sample could reasonably be considered to have a high degree of uncertainty, and so be discounted.

Finally, in a comparison of WDFW's 13 TBiOS English sole monitoring stations throughout Puget Sound, English sole from the CIRA were statistically indistinguishable from two TBiOS stations that could be considered to represent background PCB conditions in the primary (central and southern) basins of Puget Sound. The Port Madison, Anderson Island, and CIRA locations are all situated either in the main/central or southern basins of Puget Sound, and so are highly influenced by its human development, yet they are far removed from any known PCB hot spots such as Superfund sites. Hence, PCB concentrations in fish from these locations could reasonably be considered to represent background PCB levels for these Puget Sound basins, and they support the selection of CIRA as a suitable representative for that condition. The average TPCB concentration in English sole from these three background locations (12.8 ng/g wet weight) was slightly higher than the PCB recovery target (8 ng/g wet weight) adopted by the Puget Sound Partnership Vital Signs for Puget Sound Recovery. This disparity highlights Washington State's aim to reduce PCBs throughout Puget Sound, to a level lower than its current background, to protect subsistence consumers.

¹ St. Paul and Milwaukee Waterways were filled in as part of the CB/NT remedial plan, whereas Middle Waterway appears to have been largely filled in from natural processes in the period from 1984 to 2019.

Introduction and Background

The Commencement Bay Nearshore and Tideflats (CB/NT) Superfund Site in Puget Sound, Washington, USA was established in 1983 to address a wide range of contamination related to decades of chemical pollution in upland, intertidal and shallow subtidal habitats in the Commencement Bay area. The site is divided into seven operable units; the focus of this investigation is on the intertidal and shallow subtidal habitats in Operable Unit 1. An extensive remedial investigation (RI; Tetra Tech Inc., 1985(a-d)) was conducted in 1984 to determine the extent and type of contamination in Operable Unit 1, evaluate whether contamination was from then-current (1980s) or historical contamination sources (Operable Unit 5), and evaluate health risks to the public from consuming contaminated seafood. The primary objective of the 1984 RI was to define the nature and extent of contamination in the environment, including investigations of contaminants in sediment, water and tissues at concentrations that posed unacceptable risks to human health and the environment.

The extent of Operable Units 1 and 5 of the CB/NT site, original sampling site locations, and descriptions are recorded in the Superfund Record of Decision for Operable Units 1 and 5 (ROD; USEPA, 1989). The surveys conducted in 1984 established baseline status conditions for contaminants in water, sediments, invertebrate tissues, and fish tissues. Trawl tracks for the 1984 fish collections that generated tissue samples for contaminant analyses are shown in Figure 1 as red lines.

A primary objective in the ROD was to address human exposure pathways associated with contaminated marine sediments, and to focus source controls and sediment remedial actions to reduce contaminants in seafood originating from upland sources in the CB/NT. Section 7 of the ROD identified PCBs as the sole human health contaminant of concern for Operable Unit 1 of the CB/NT. The human health risk assessment conducted for Operable Unit 1 calculated the risk to humans consuming PCB-contaminated fish from the CB/NT site, and established advisories to limit consumption to protect human health. It further stated the fish tissue recovery objective for the average PCB concentration measured in English sole from CB/NT to be equal to or less than 36 µg/kg (or ng/g ww), wet weight². This target concentration represented a background reference condition for PCBs (measured as a summation of detected Aroclors, hereafter referred to as Total Aroclors) in English sole sampled from the Carr Inlet reference area (CIRA), a nearby embayment with no known or suspected local PCB inputs. This document relates solely to PCBs measured in English sole muscle (fillet with skin removed) tissue, and whether tissue PCB concentrations have declined sufficiently (below the CIRA tissue concentrations) to determine if the cleanup objective for human consumption of fish has been attained and further site restrictions for fish consumption are warranted.

Comparisons of PCBs through time, especially in environmental media, are only valid when changes in analytical methods are considered and accounted for (Butcher et al. 1996). Analytical methods for PCBs in fish tissue have changed substantially over the 34 years between the baseline and current study, highlighting a need to consider the validity of the original measurements and their comparability with current methods. Some of the key factors include the methods for extracting organics from tissue (both solvent type and method), the limit of quantitation, accuracy and precision, and other quality assurance/quality control parameters. The method used to measure Aroclors in 1984 was not explicitly described in the RI, so there is substantial uncertainty regarding its comparability with the current Aroclor methods applied to the 2019 samples collected in this study (EPA Method 8082A). For these

² Our calculation of this target from raw data in the RI (Tetra Tech Inc. 1984d) resulted in an arithmetic average concentration of 38 ng/g ww

reasons, it was necessary to also resample the CIRA in 2019 for comparison to the CB/NT site data rather than comparing the site data to the older reference concentration.

Using modern Aroclor methods to measure total PCBs in fish tissues or other environmental media can also be problematic in ways that could significantly affect the interpretation of data generated from English sole in this study. PCB congener proportions in environmental samples, especially biota, can differ markedly from their original proportions in Aroclors (Sather et al. 2003), potentially creating problems in accurately quantitating PCBs using Aroclor methods. Complex environmental weathering processes can change the relative abundance of congeners, potentially yielding both false negative results for Aroclors (Wischkaemper et al. 2013), or overestimation bias (Sather et al., 2003). EPA recommended as early as 2001 that Aroclor analysis of environmental samples be used only to confirm relatively recent contamination, rather than older, more potentially weathered PCBs (Beliveau 2001). However, Aroclor data are still used to determine human health risk because the cancer slope factor used to estimate risk is based on Aroclors.

For these reasons, WDFW and EPA agreed to additionally measure PCBs using current congener-based methods to estimate total PCBs (hereafter termed TPCB) in the 2019 tissue samples in both CB/NT and the CIRA. These measurements, combined with TPCB measured in English sole and other species from 13 long-term monitoring index sites in Puget Sound (West et al., 2017; Puget Sound Partnership, 2022a-d) provide context for interpreting 2019 CB/NT PCB location patterns. Such comparisons are not only useful to understand whether the CB/NT remedies have resulted in sufficient recovery to meet the ROD tissue objective, but also to understand how they relate to current background conditions and whether they meet current Washington State recovery targets for Puget Sound.

Methods:

Field Sampling

The CB/NT Remedial Investigation (RI, Tetra Tech, Inc., 1985a-d) reported PCB concentrations from seven industrial waterways and three shoreline locations along the western shoreline of Commencement Bay, as well as two locations in the Carr Inlet reference area in 1984 (RI, Figure 2.6). Separate locations were sampled within the three largest waterways: three locations each in Blair and Hylebos (head, middle and mouth), and two locations in Thea Foss Waterway (head and mouth). WDFW successfully occupied all the original 1984 trawl locations except Milwaukee, Middle and St. Paul Waterways, all of which had been mostly filled in between 1984 and 2019. When possible, English sole were sampled offshore along the shoreline fronting the location of these three former waterways. The comparison of locations between the sampling years was accomplished by plotting the 2019 trawl tracks using GIS-collected polygons during the trawling (yellow lines for CB/NT efforts and cyan lines for TBIOS long-term monitoring efforts), with trawl lines transcribed by eye from Figure 2.6 of the RI (red lines; Figure 1- industrial waterways; Figure 2– three shoreline locations along the western shoreline of Commencement Bay; Figure 3 – CIRA).

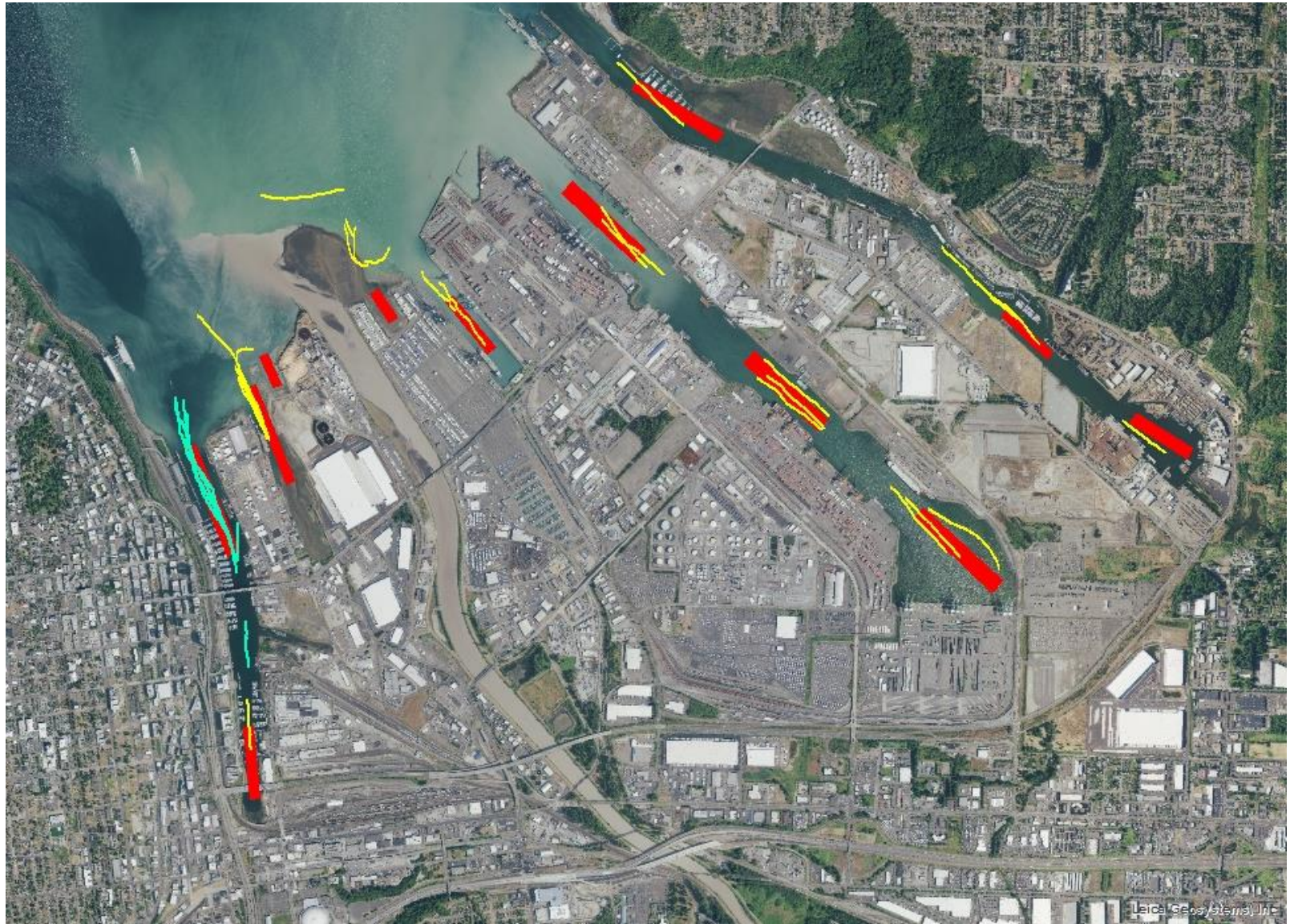


Figure 1. Location of 1984 (red lines) and 2019 trawl (yellow lines) sampling efforts to collect English sole in CB/NT industrial waterways. Cyan lines indicate trawl tracks from the 2019 TBiOS long-term monitoring effort.

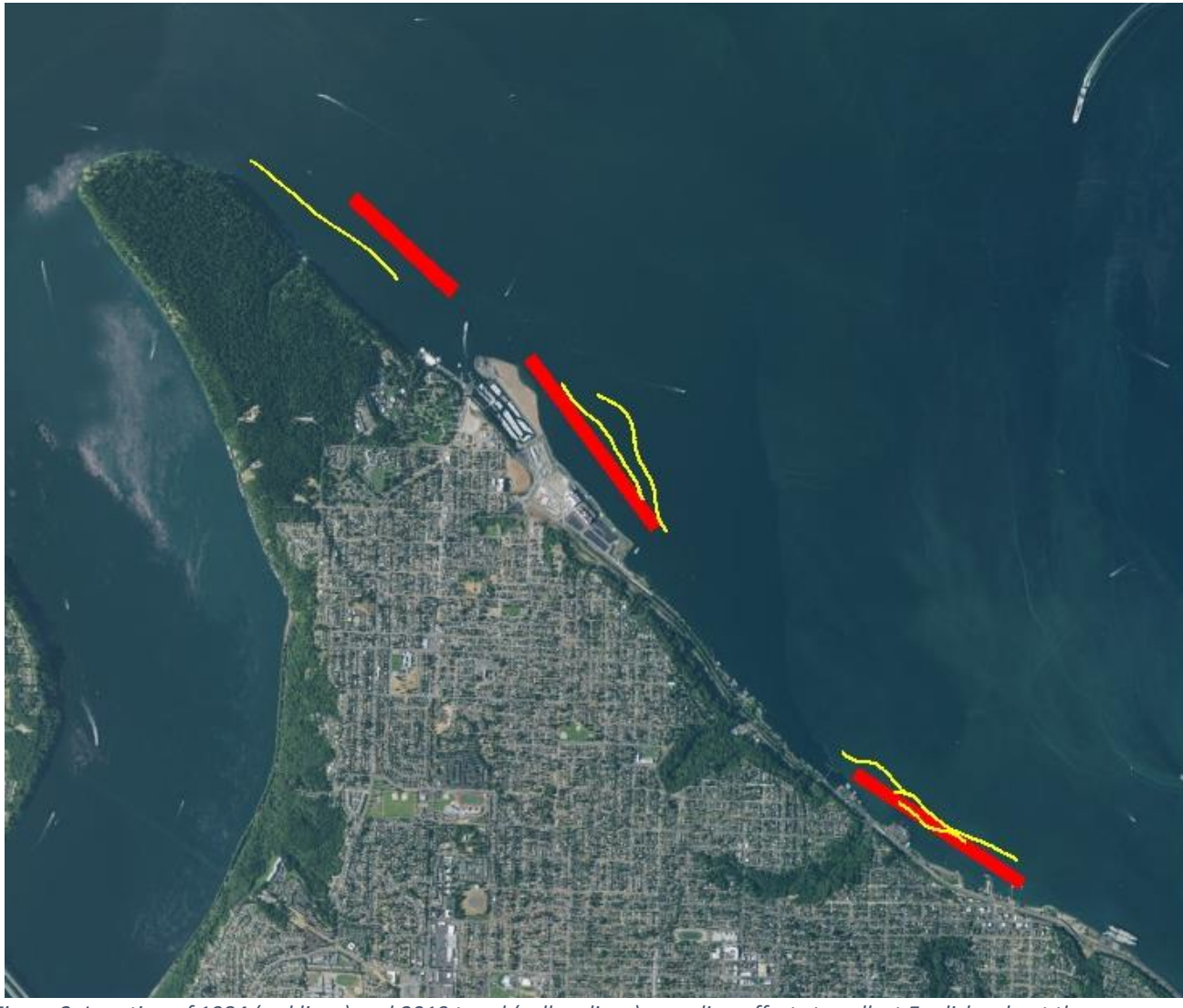


Figure 2. Location of 1984 (red lines) and 2019 trawl (yellow lines) sampling efforts to collect English sole at three shoreline locations along the western shoreline of Commencement Bay.

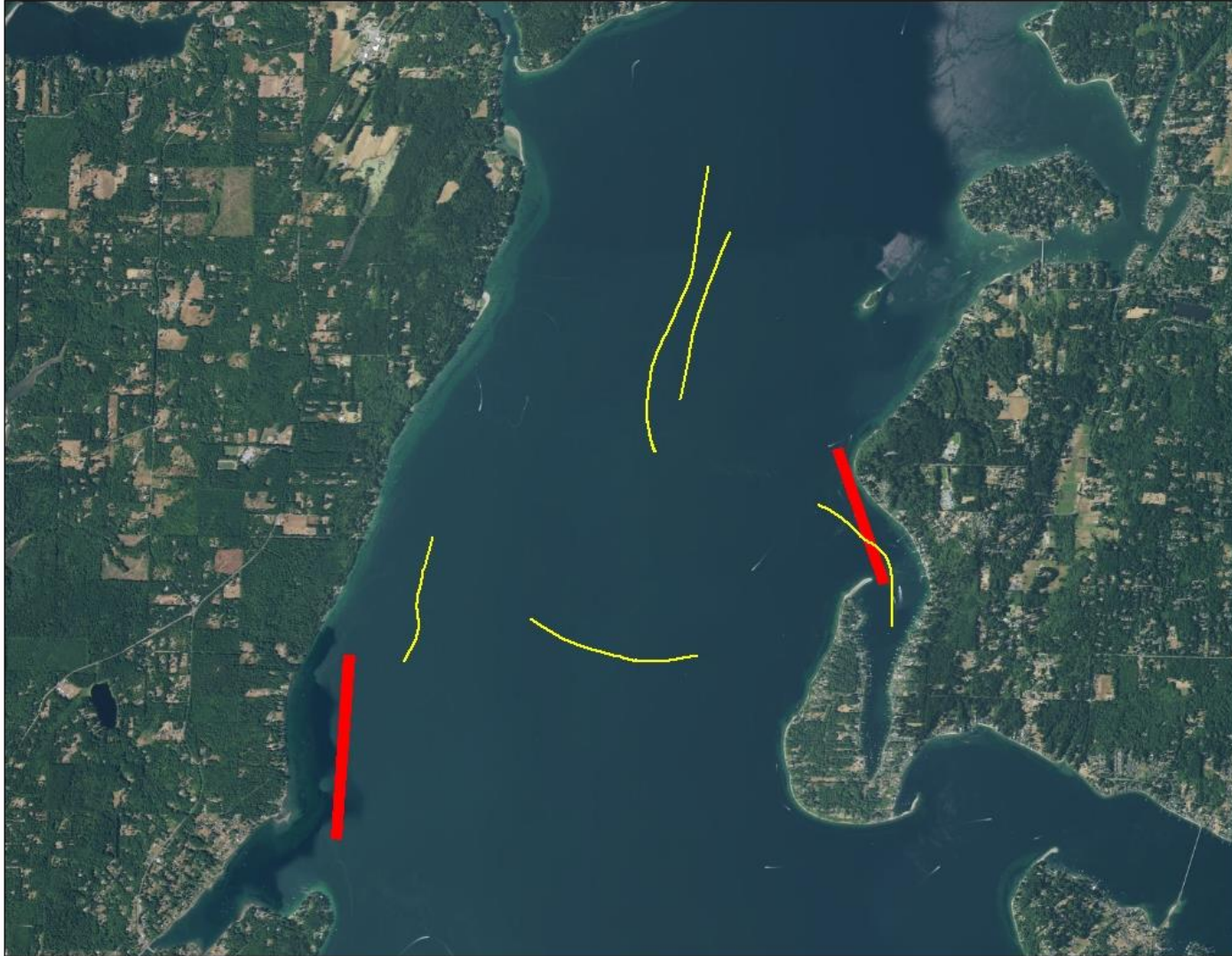


Figure 3. Location of 1984 (red lines) and 2019 trawl sampling (yellow lines) efforts to collect English sole from the Carr Inlet Reference Area (CIRA).

WDFW collected fish and processed tissues in 2019 using the 1984 methods. The 1984 sampling methodology for English sole is documented in the 1984 RI (section 2.7.1.1 and 2.7.1.2.) and remains similar to WDFW's current TBIOS status and trends monitoring methods (Washington Department of Fish and Wildlife in prep (a)). Any apparent or suspected deviations are noted herein.

English sole were collected over a four-day period from 7/15/2019 to 7/18/2019 using a bottom trawl similar to the one used in 1984. Only fish greater than 23cm in total length were used for both studies. Up to five fish were selected for tissue analysis from each location, held on ice, and transported to a land-based lab for later tissue resection. It appears the 1984 fish were resected without having been frozen, whereas the 2019 fish were frozen in the lab to -20 deg. F until resection was conducted. The sex of each fish was determined and reported in 2019, but not reported in 1984.

Resecting muscle tissue from frozen-thawed whole fish is the current method used by WDFW. It is considered equivalent to processing fish held on ice and not frozen, for analysis of PCBs and other persistent organic pollutants in muscle tissue. The 1984 whole fish were cleaned with tap water prior to being filleted, and muscle tissue was subsequently scraped free of the fish skin. The 2019 fish were processed by first removing the skin, and then resecting muscle tissue from the carcass. Both methods are considered acceptable and equally protective of the sample tissue. All instruments in both studies were solvent-cleaned with methylene chloride (1984) or isopropyl alcohol (2019), and subsequently rinsed with deionized water before resecting tissues, and resected tissues were placed in glass jars (unknown type in 1984, and certified solvent-cleaned I-Chem series 2 jars in 2019). Work surfaces in both studies were similarly cleaned between samples or covered with pre-cleaned foil (2019). It is unclear how tissues were stored in the 1984 study prior to chemical analysis, but it is presumed they were frozen and held at -20 deg. F, as they were in 2019. All 2019 tissue samples were homogenized using electric grinders with stainless steel blades, to a paste-consistency. There is no description regarding how the 1984 samples were homogenized but it is presumed a similar method was used.

All tissue samples analyzed in 1984 were individual fish. This was the case for the 2019 study as well, except for the CIRA, where composite samples (5 fish per composite) were created to increase the number of fish to represent the larger Carr Inlet area. Eighty-five field tissue samples from 15 locations representing four industrial waterways, the shoreline area offshore of three mostly in-filled industrial waterways, three areas along the Commencement Bay western shoreline, and the CIRA were created using this procedure (Table 1). Multiple locations were sampled within the three long waterways, Hylebos, Blair and Thea Foss Waterways, to potentially distinguish between head, middle, and mouth areas. Sample size (n), minimum, maximum and arithmetic mean values for Total Aroclors and TPCB are reported in Table 1.

Chemical Analyses: 2019 Aroclors

Resected English sole muscle tissues were homogenized at the WDFW TBIOS laboratory in Olympia, WA, using standard operating procedures for tissue homogenization (Washington Department of Fish and Wildlife, in prep (b)). Samples were then split, with aliquots sent to separate labs for Aroclor and PCB congener analysis. Aroclors from the 85 CB/NT and CIRA samples collected in 2019 were analyzed at the USEPA Region 10 Laboratory, 7411 Beach Dr. East, Port Orchard, WA, 98366 in January and February 2020 using the most current standard operating procedures from EPA Method 8082A at the time. Samples were extracted using EPA method 3541 (automated soxhlet extraction) with EPA cleanup methods 3665A (sulfuric acid) and 3620C (florisil). Seven Aroclor mixtures were quantitated (Aroclors

1016, 1221, 1232, 1242, 1248, 1254, and 1260). Of the seven Aroclors quantitated, only Aroclor 1254 and 1260 were detected.

Aroclor 1254 was detected in 36/85 samples (42 percent) with concentrations ranging from 11 to 130 ng/g wet weight, and Aroclor 1260 was detected in 71/85 (86 percent) of tissue samples with concentrations ranging from 12 to 120 ng/g ww. The reporting limit for all non-detected Aroclors ranged from 8.7 to 9.6 ng/g wet weight. All quality assurance goals and quality control measures were achieved. Aroclors were never detected in any of the drying agent blanks. Total Aroclors were calculated as a sum of detected Aroclor concentrations. In cases where no Aroclor was detected (14 samples), PCBs were reported as the non-detect concentration for a single Aroclor (in all such cases the reporting limit for Aroclors 1254 and 1260 were identical).

Chemical Analyses: 2019 Congener-PCBs and Lipids

PCB congener analyses were conducted on tissue aliquots at NOAA's Northwest Fisheries Science Center (NWFSC, Seattle, WA). Tissue samples were analyzed using GC/MS with accelerated solvent extraction (ASE) according to Sloan et al. (2014). In brief, this method comprised three steps; 1) ASE of tissue using methylene chloride (MeCl), 2) a two-step cleanup of the MeCl extract by silica/aluminum columns and size-exclusion high-performance liquid chromatography (SEC HPLC) to remove lipids and other biogenic compounds, and 3) quantitation of PCBs using GC/MS with selected-ion-monitoring (SIM). The ASE provided an extract that was used for analyte recovery and gravimetric lipid evaluation. Accuracy of the instrument was improved by including chemical ionization filaments (used to increase source temperature), a cool on-column injection system in the GC, and a guard column before the analytical column. Point-to-point calibration was used improve data fit over the full range of GC/MS calibration standards (Sloan et al. 2014). Forty-seven PCB congeners were quantitated in all samples.

Total congener-PCBs (TPCB) were estimated using a slightly modified algorithm from Lauenstein et al. (1993) as described in West et al. (2017), and reprinted here with slight modifications to the narrative; TPCBs were estimated using a simple algorithm based on a subset of 17 PCB congeners commonly detected in marine organisms, representing homologs containing three to ten chlorine atoms (IUPAC numbers 18, 28, 44, 52, 95, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, and 209) wherein the sum of detected values for these 17 congeners was multiplied by 2 to estimate TPCB. This algorithm was developed for the US nation-wide toxics monitoring program from empirical data (Lauenstein and Cantillo 1993), as a cost-effective way for high-sample-volume monitoring programs to estimate total PCBs from a small subset of congeners using relatively inexpensive gas chromatography/mass spectrometry analytical techniques. Total PCBs in that program were estimated by Lauenstein and Cantillo (1993) using 18 congeners (those above, plus PCB 8); however, PCB 8 was not quantitated in the current study, so the algorithm employed herein summed only 17 congeners. Although PCB 8 was detected in 30 samples of fish tissue in another study of Puget Sound fish using high-resolution GC/MS methods (West et al. 2011), concentrations were extremely low, ranging from 0.0014 to 0.012 ng/g wet wt, suggesting it would be undetectable by the methods used in this study. Lauenstein and Cantillo's (1993) $2 \times \sum 18 \text{ PCBs}$ provided excellent agreement with total PCBs estimated from a total chlorination method in their study, and $2 \times \sum 17 \text{ PCBs}$ in the current study agreed well with total PCBs estimated from the sum of 209 congeners measured by high resolution methods (linear regression of $\sum 209$ congeners by $2 \times \sum 17 \text{ PCBs}$, $r^2=0.997$, $p=0.003$, slope coefficient=0.991, intercept not significant, $p=0.892$) for a subset of five English sole and herring from West et al. (2017).

Data Analysis Statistics

All PCB location comparisons were made using Analysis of Variance (ANOVA) models with a Holm-Sidak multiple comparison test or Holm-Sidak multiple comparison test versus a control group (using CIRA as PCB concentrations as the control), with α set to 0.05. Although all statistical comparisons were made on log-10 transformed data to help reduce heteroscedasticity for the ANOVAs, results for all data are presented using arithmetic metrics. Time trends were modeled using a general linear model of log-10-transformed PCB concentration with year as the independent factor. Linear regression of TPCB and Total Aroclors measured in sample splits was conducted using a general linear model on log-10-transformed data to predict one from the other.

WDFW English Sole Monitoring Data

WDFW's long-term English sole monitoring data, used herein for comparison with CB/NT results, was taken from sites sampled in 2019, recently reported on the Puget Sound Partnership's Toxics in Fish Vital Sign (Puget Sound Partnership 2022a). WDFW's monitoring sites were designed to provide wide coverage across the entire Puget Sound with site conditions ranging from highly industrialized or urbanized locations to remote locations far removed from any known or suspected PCB sources (Figure 4, adapted from West et al. 2017). Location latitudes and longitudes for the average position of the midpoint of all trawl tracks (centroids) are presented in Table 1. All field and laboratory methods used in the TBIOS monitoring program are consistent with the current CB/NT study except for the number of fish used in the preparation of tissues samples. Whereas the current study analyzed typically five individual fish from each CB/NT location, TBIOS monitoring creates up to 6 composite samples from each monitoring location, each comprising up to 20 randomly selected individuals. Hence, although TBIOS monitoring is not necessarily statistically more robust (six analytical samples versus five), it likely better represents the true population contaminant condition.

Table 1. Total Aroclor and TPCB concentrations (ng/g ww) in English sole muscle tissue from 15 CB/NT sampling locations, the Carr Inlet Reference Area (CIRA) and WDFW's 13 TBIOS English sole monitoring stations from 2019 (see Figure 4 for TBIOS site locations). CB/NT samples were collected over a four-day period from 7/15/2019 to 7/18/2019.

| Site | Centroid of Trawl Tracks | | Total Aroclors | | | | TPCB | | | |
|--|--------------------------|------------|----------------|------|-----|-----|------|------|-----|-----|
| | Latitude | Longitude | n | Mean | Min | Max | n | Mean | Min | Max |
| <u>CB/NT Project Locations³</u> | | | | | | | | | | |
| Carr Inlet Ref. Area (b) | 47.30597 | -122.71185 | 6 | 39 | 24 | 47 | 6 | 21 | 14 | 33 |
| Old Town | 47.28133 | -122.47326 | 5 | 17 | 13 | 27 | 5 | 15 | 9.8 | 23 |
| Ruston | 47.30182 | -122.50048 | 5 | 14 | 9.3 | 17 | 5 | 13 | 9.2 | 20 |
| Pt Defiance | 47.31511 | -122.52646 | 5 | 25 | 9.3 | 67 | 5 | 17 | 4.9 | 30 |
| Thea Foss WW (head) | 47.24691 | -122.43241 | 5 | 35 | 16 | 50 | 5 | 40 | 20 | 55 |
| Thea Foss WW (mouth ⁴) (c) | 47.25821 | -122.43541 | 5 | 63 | 14 | 84 | 5 | 63 | 16 | 85 |
| Middle WW | 47.26386 | -122.43259 | 7 | 18 | 9.3 | 39 | 7 | 21 | 5.4 | 39 |
| Milwaukee WW | 47.27235 | -122.42463 | 6 | 10 | 9.2 | 13 | 6 | 13 | 7.8 | 20 |
| Sitcum WW | 47.26935 | -122.41752 | 5 | 15 | 9.5 | 19 | 5 | 17 | 5.3 | 28 |
| Blair WW (head) | 47.25856 | -122.37987 | 9 | 43 | 9.1 | 216 | 9 | 44 | 14 | 230 |
| Blair WW (middle) | 47.26479 | -122.39080 | 5 | 16 | 13 | 19 | 5 | 17 | 12 | 23 |
| Blair WW (mouth) | 47.27277 | -122.40389 | 5 | 18 | 9.4 | 33 | 5 | 18 | 8.4 | 28 |
| Hylebos WW (head) | 47.26320 | -122.36359 | 7 | 68 | 32 | 189 | 7 | 73 | 33 | 210 |
| Hylebos WW (middle) | 47.27024 | -122.37540 | 5 | 147 | 115 | 201 | 5 | 160 | 110 | 230 |
| Hylebos WW (mouth) | 47.28062 | -122.40246 | 5 | 50 | 41 | 59 | 5 | 47 | 38 | 56 |
| St. Paul WW | not sampled | | - | -- | -- | -- | - | -- | -- | -- |
| <u>TBIOS Stations</u> | | | | | | | | | | |
| Anderson Island (a) | 47.15764 | -122.66902 | - | -- | -- | -- | 6 | 13 | 11 | 14 |
| Thea Foss WW (mouth) (c) | 47.25821 | -122.43541 | - | -- | -- | -- | 3 | 100 | 79 | 120 |
| Sinclair Inlet (d) | 47.54978 | -122.63655 | - | -- | -- | -- | 6 | 43 | 34 | 51 |
| Eagle Harbor (e) (continued) | 47.62054 | -122.50893 | - | -- | -- | -- | 6 | 46 | 34 | 59 |
| West Point (f) | 47.66865 | -122.43847 | - | -- | -- | -- | 4 | 33 | 32 | 34 |

³ Lower case letters refer to locations on map, Figure 4

⁴ TBIOS and CB/NT fish samples for Thea Foss Waterway (mouth) location were taken from the same trawling effort

Table 1 continued.

| Site | Centroid of Trawl Tracks | | Total Aroclors | | | | TPCB | | | |
|--------------------------------|--------------------------|------------|----------------|------|-----|-----|------|------|-----|-----|
| | Latitude | Longitude | n | Mean | Min | Max | n | Mean | Min | Max |
| Duwamish River (g) | 47.55855 | -122.34451 | - | -- | -- | -- | 5 | 430 | 300 | 510 |
| Seattle Waterfront (h) | 47.60803 | -122.35069 | - | -- | -- | -- | 6 | 82 | 28 | 140 |
| Seattle (Myrtle Edwards) (i) | 47.62020 | -122.37511 | - | -- | -- | -- | 4 | 160 | 130 | 210 |
| Hood Canal (j) | 47.83678 | -122.64010 | - | -- | -- | -- | 6 | 4.4 | 2.8 | 5.3 |
| Port Madison (k) | 47.73089 | -122.50601 | - | -- | -- | -- | 6 | 13 | 8.2 | 18 |
| Everett (Port Gardner Bay) (l) | 47.98397 | -122.24483 | - | -- | -- | -- | 6 | 21 | 15 | 28 |
| Vendovi Island (m) | 48.64471 | -122.64358 | - | -- | -- | -- | 6 | 1.3 | 1.0 | 1.6 |
| S. Strait of Georgia (n) | 48.86126 | -122.96035 | - | -- | -- | -- | 3 | 2.2 | 1.4 | 3.0 |

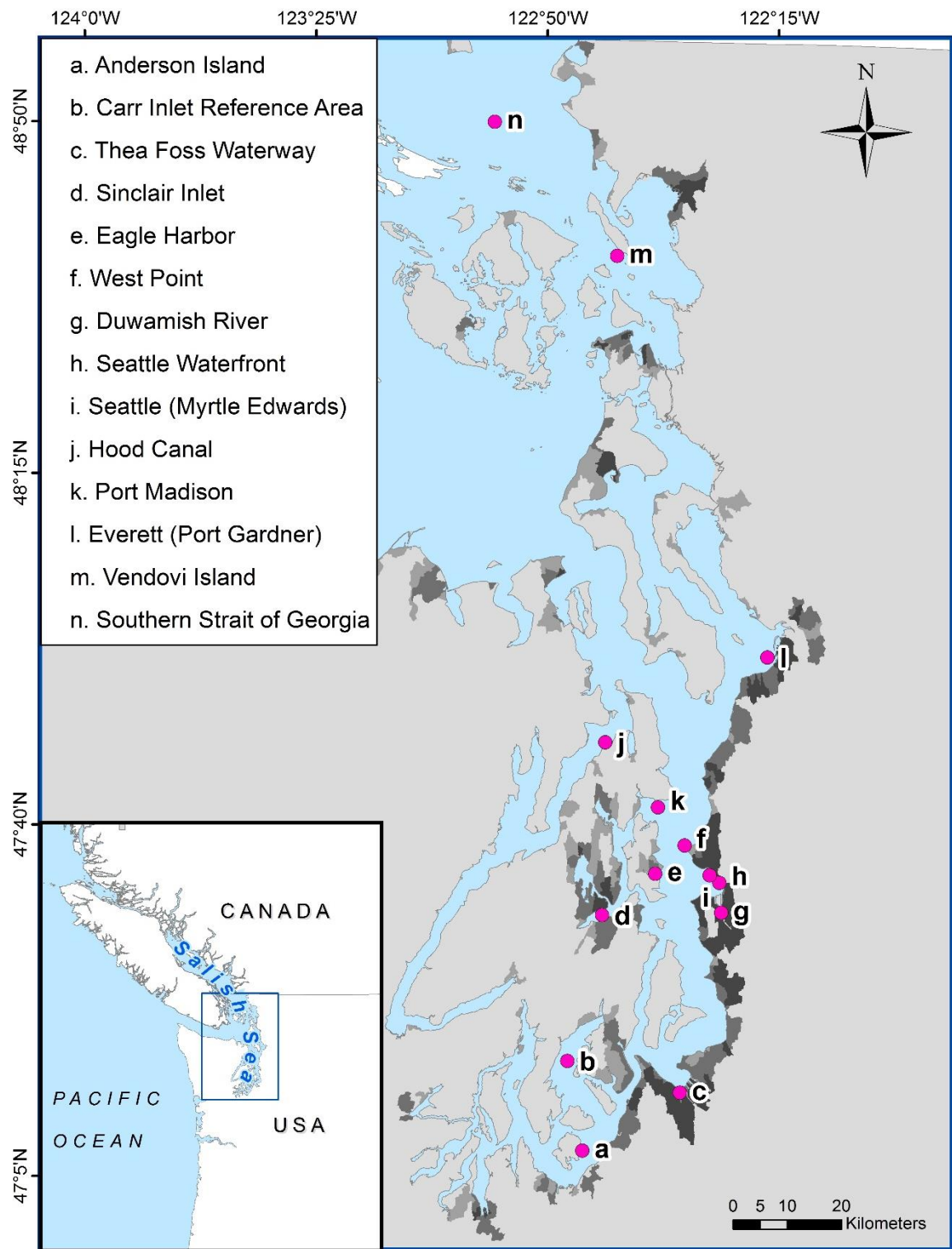


Figure 4. Location of WDFW's TBIOS monitoring sites for English sole. Gray shading indicates the intensity of land-development, as represented by the percentage of land covered by impervious surfaces, adapted from West et al., (2017).

Results and Discussion

Aroclor Evaluation of CB/NT Remediation

The primary goal for EPA with this sampling event was to determine whether the average PCB concentration in fish (English sole) tissue following sediment cleanup at the CB/NT site had decreased to the concentrations measured in English sole from the Carr Inlet Reference Area (CIRA). Based on this comparison, the CB/NT tissue PCB objective has apparently been achieved at the spatial scale of Operable Unit 1; mean Total Aroclors measured in 79 pooled CB/NT fish tissue samples taken in 2019 (41 ng/g ww) were statistically indistinguishable from the mean concentration in CIRA English sole taken in 2019 (38 ng/g wet wt; ANOVA of log-10-transformed Total Aroclors by year; $p=0.89$). Although this primary goal was met at spatial scale of the entire Operable Unit 1, the following analyses show PCB contamination in fish tissue significantly above the CIRA reference persisted in two industrialized waterways in 2019 (Figure 5 -- Hylebos and Thea Foss Waterways), and a third (Blair Waterway) exhibited contamination sufficient to warrant concern. A deeper analysis of PCB contamination at these three locations considering both Total Aroclors and TPCB is presented below.

Of the remaining CB/NT locations, all samples from Sitcum Waterway exhibited Total Aroclor concentrations below the CIRA concentration (9.5-19 ng/g ww). Milwaukee, St. Paul, and Middle were substantially filled-in either naturally or intentionally as part of the remedial plan, eliminating most of the waterway habitat that had been sampled in 1984. However, English sole taken at nearshore locations fronting Milwaukee and Middle Waterways (bracketing the location of the former St Paul Waterway – Figure 1) exhibited Total Aroclor concentrations below the CIRA reference (9.2-39 ng/g for both locations combined – Table1), supporting a conclusion that the remediation of Sitcum Waterway and this shoreline area were successful. English sole from the three Commencement Bay western shoreline locations (Pt. Defiance, Ruston, and Old Town) did not exhibit elevated Aroclor concentrations in 1984 relative to the CIRA (Tetra Tech 1985 a-d) and were not remediated for PCBs. Aroclor concentrations in all 2019 samples from these three shoreline locations were also substantially below the CIRA reference, ranging from 9.3 to 27 ng/g ww, except for one Pt Defiance sample with a concentration of 67 ng/g ww (resulting in large confidence intervals; Figure 5).

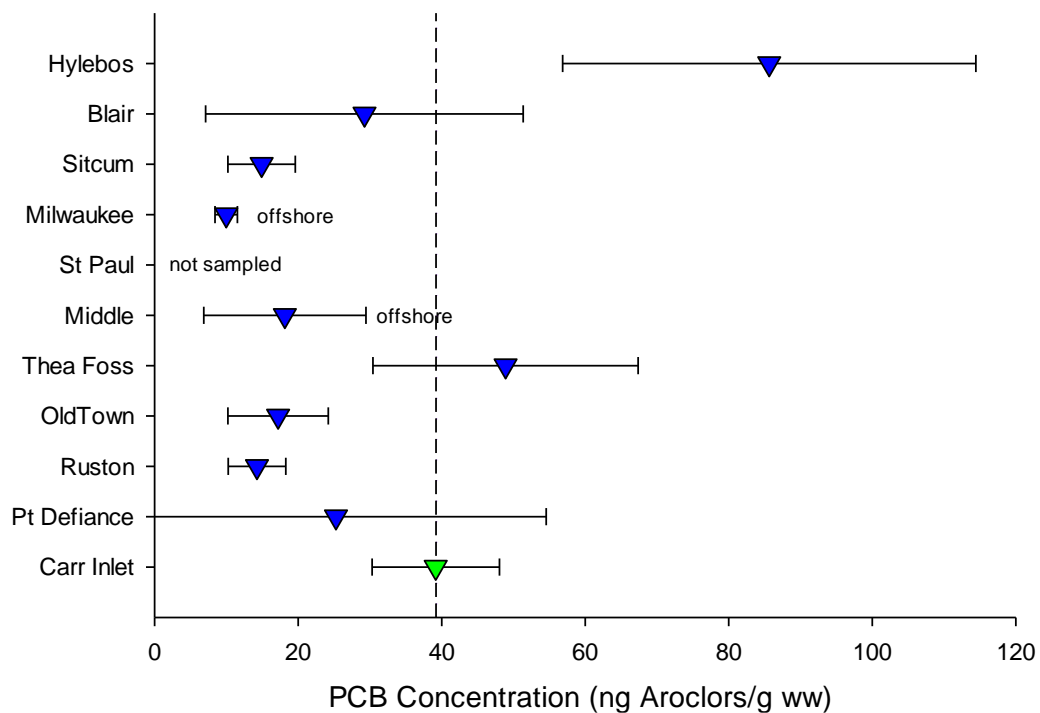


Figure 5. Comparison of Total Aroclors in English sole sampled in 2019 from six waterway locations and three open shoreline locations in CB/NT (blue triangles) with Total Aroclors in English sole sampled from the Carr Inlet reference area (CIRA, green triangle) in that year. Triangles indicate the arithmetic mean PCB concentration, with 95 percent confidence intervals. The vertical dashed line indicates the CIRA PCB concentration of 39 ng/g ww.

A PCB-congener Approach to Evaluating CB/NT Remediation

Analysis of congener PCBs on the same samples reported above for Aroclors supports and appears to clarify the Aroclor patterns with greater statistical certainty (Figure 6). The mean concentration of TPCB in English sole from both Hylebos (92 ng/g ww) and Thea Foss⁵ (63 ng/g ww) Waterways were significantly greater ($p=0.005$ and 0.041) than the CIRA TPCB concentration (21 ng/g ww) whereas PCBs in English sole from all other CB/NT locations were statistically indistinguishable from the CIRA ($p>0.74$; ANOVA of log₁₀-transformed PCB data, *post hoc* pairwise comparisons using the Holm-Sidak method, with CIRA as a control). Blair Waterway was the only other location (besides Thea Foss and Hylebos) exhibiting a mean PCB concentration greater than the CIRA, albeit not significantly so.

The additional statistical significance conferred by the TPCB congener analysis in this comparison appears to be related to the lower TPCB concentration measured in fish from the CIRA (mean of 21 ng/g ww) compared to the Total Aroclor CIRA mean of 39 ng/g ww. This is consistent with the Total Aroclor:TPCB relationship illustrated in Figure 7, which shows a comparison of these two analytical procedures for all 85 sample splits. Whereas Total Aroclors and TPCB analyses generally returned similar concentrations in the upper end of the concentration range, a substantial departure occurred at the lower end (approximately below 40 ng/g ww) with Total Aroclors overestimating PCBs relative to TPCB.

⁵ CB/NT Thea Foss data combined with TBIOS Thea Foss data

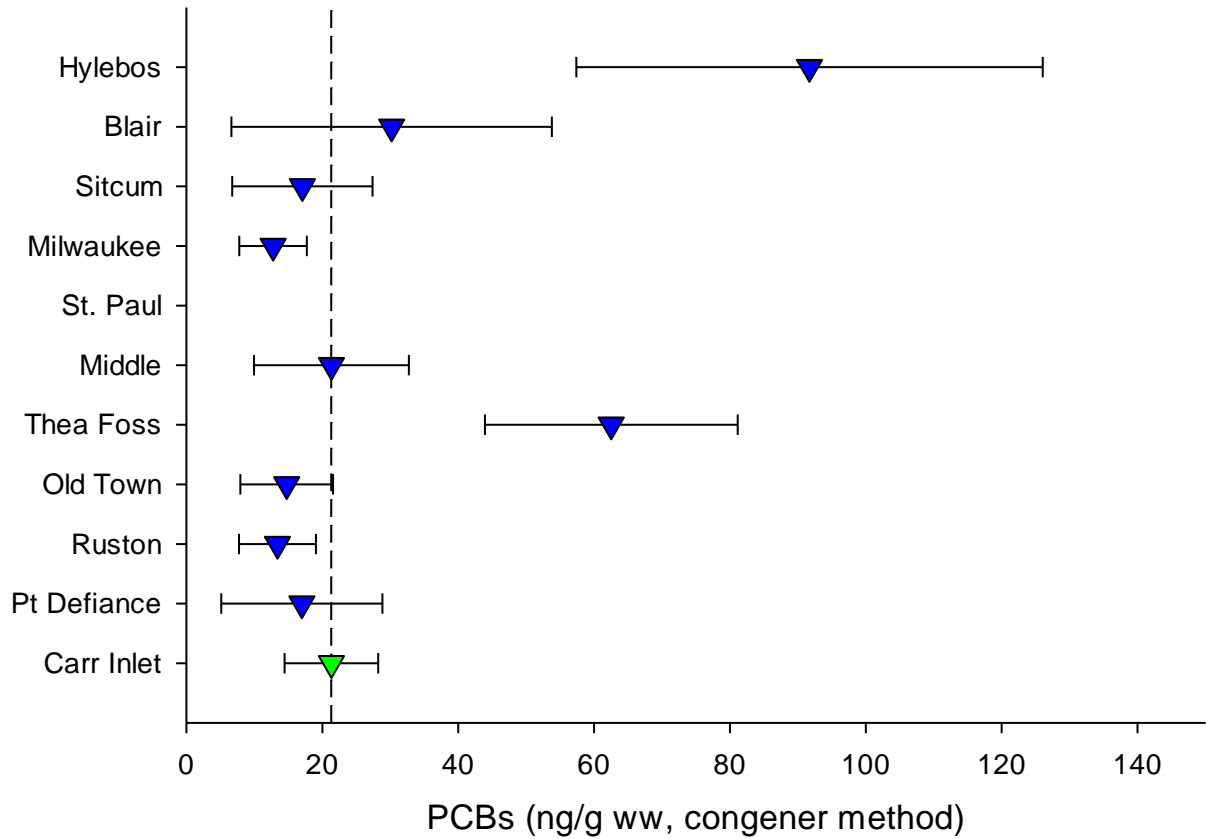


Figure 6. Comparison of PCBs measured by congeners in English sole sampled in 2019 from six waterway locations and three open shoreline locations in CB/NT (blue triangles) with PCBs in English sole sampled from the Carr Inlet reference area (CIRA, green triangle) in that year. Triangles indicate the arithmetic mean PCB concentration, with 95 percent confidence intervals. For reference, the vertical dashed line indicates the CIRA PCB concentration of 21 ng/g ww.

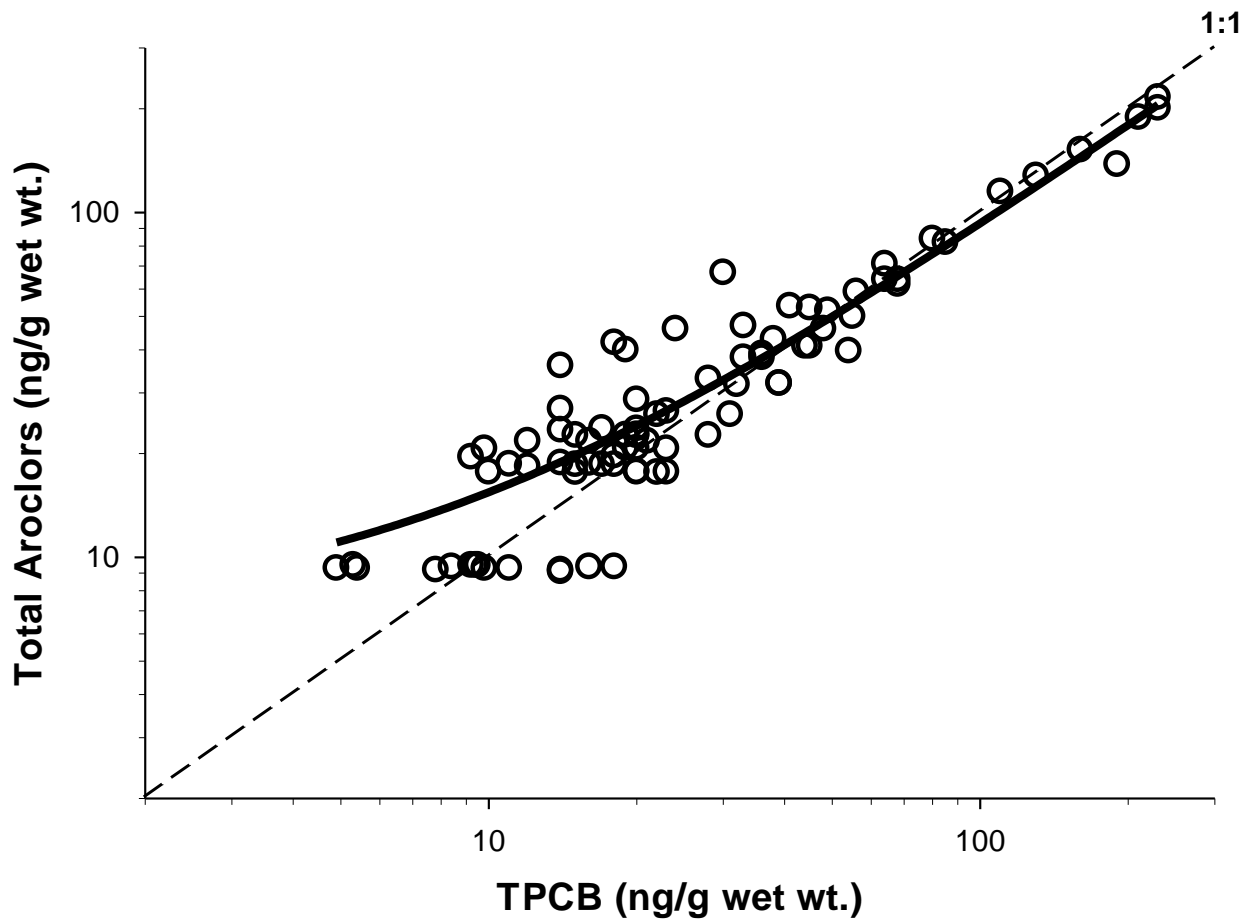


Figure 7. Comparison of Total Aroclor and TPCB concentrations measured from splits taken from all 85 tissue samples. The dashed line represents a one-to-one relationship. The solid line represents the predicted relationship between these two measurements based on a linear regression of log-10-transformed data for Total Aroclors and TPCB (adjusted $r^2=0.94$, with intercept and slope coefficients of 6.8 and 0.86 ($p<0.0001$ for both)).

A Comparison of CB/NT with English Sole from Other Puget Sound Locations

Further analysis of sub-locations (head, middle and mouth) for the three waterways containing high PCB concentrations helps pinpoint where contamination has persisted. Moreover, considering WDFW's long-term TBiOS monitoring sites in addition to the CB/NT sampling locations provides a context for comparing PCB conditions in CB/NT with the rest of Puget Sound. This includes other non-urban locations similar to Carr Inlet representing reference or background conditions, as well as other Puget Sound locations considered to be highly contaminated with PCBs. The combination of all CB/NT and TBiOS monitoring sites sampled in 2019 resulted in 155 composite or individual muscle tissue samples for TPCBs, representing 1335 English sole from 27 sites throughout Puget Sound. Of those, 12 were WDFW monitoring stations outside of CB/NT, 14 were from areas within the CB/NT and one was the CIRA (Table 1). One location, the Thea Foss Waterway mouth, was sampled for both TBiOS and CB/NT purposes from the same field effort; the Thea Foss Waterway mouth also serves as a long-term TBiOS monitoring station.

Three CB/NT sub-locations stand out as exhibiting high PCB contamination, similar to locations with the greatest PCB contamination in Puget Sound; the Hylebos middle and head locations, and Thea Foss mouth location (Figure 8). The average concentration of TPCBs in English sole from the Hylebos middle location was statistically indistinguishable from the English sole sampled in the Duwamish River in 2019, the most highly PCB-contaminated of the 13 long-term TBIOS monitoring stations. Moreover, although the average TPCB concentration in Hylebos head and Thea Foss mouth locations were statistically lower than the Duwamish River, they were indistinguishable from two other highly contaminated monitoring sites, the Seattle Waterfront and Seattle (Myrtle Edwards) stations (ANOVA of log-10-transformed TPCB by location, Holm-Sidak post hoc pairwise multiple comparison procedure, $\alpha = 0.05$). Average TPCB in English sole from three other locations, Thea Foss head, Hylebos mouth, and Blair head, locations exhibited TPCB concentrations roughly twice the concentration of fish from the CIRA (Table 1, Figure 8), however these differences were not statistically significant (same ANOVA described above).

English sole from the Blair Waterway head location exhibited one of the two greatest TPCB concentrations (230 ng/g ww) in the entire CB/NT study. Seven of 19 samples from all three Blair Waterway sub-locations combined (37%), and four of nine samples (44%) from the Blair Waterway head exhibited TPCB above the average CIRA concentration, suggesting continued contamination in that waterway. The distribution of high values points to the head of the waterway as the likely source.

English sole from all other CB/NT locations; Sitcum Waterway, the three nearshore areas fronting the former Milwaukee, St. Paul, and Middle Waterways, and the 3 western shoreline sites of Commencement Bay (Old Town, Ruston, and Pt. Defiance) appear to have met the intent of the CB/NT recovery. Their average PCB concentrations in 2019 were statistically indistinguishable from not only the CIRA, but also from two other TBIOS monitoring locations that could also be considered to represent Puget Sound central basin (Port Madison) and southern basin (Anderson Island) background PCB conditions (Figure 8). Port Madison and Anderson Island stations occur in basins impacted by human population and land-development, however they are relatively far removed from known or suspected PCB hot spots or sources. The average TPCB concentration of English sole from these three background reference locations (CIRA, Port Madison, and Anderson Island) was 12.8 ng/g wet wt (calculated from raw data summarized in Table 1)

PCB levels in CB/NT English sole compared to Washington State's Puget Sound recovery targets

USEPA and Washington State's Puget Sound Partnership (PSP) have worked together for over ten years to develop a Puget Sound Comprehensive Conservation and Management Plan to achieve Puget Sound ecosystem health recovery goals. Puget Sound is designated as an Estuary of National Significance under USEPA's national Estuary Program. A Puget Sound Management Conference, comprising staff from USEPA, PSP, and a broad coalition of local stakeholders have developed long-term strategies and plans to address water quality and ecosystem health issues (including PCB contamination). The most current Puget Sound ecosystem recovery goals and strategies are being reviewed for an upcoming four-year period in a document titled "*2022-2026 Action Agenda: An update to the Puget Sound recovery plan for protected habitat, thriving species, and healthy people*" (Puget Sound Partnership, 2022c)

Under this Puget Sound Action Agenda recovery plan, and as a member of the Puget Sound Ecosystem Monitoring Program (PSEMP), WDFW's TBIOS is responsible for monitoring the status and trends of toxic contaminants in select indicator species in Puget Sound, including PCBs in English sole, Pacific herring, various salmon species, and mussels. All of the TBIOS English sole data reported in Figure 8,

most of the time trends data in Figure 9, mussel data in Figure 10, and salmon data reported in Figure 11 were created from these efforts, with data sources cited herein from specific reports or from the Puget Sound Partnership's Toxics in Aquatic Life Vital Signs web site (Puget Sound Partnership, 2022a).

The Toxics in Aquatic Life Vital Sign describes Washington State's recovery goals for each species and each major contaminant class, including PCBs, for two time horizons – 2030 and 2050. For English sole these recovery targets are:

- By 2030, 95% of the samples gathered across Puget Sound habitats exhibit a declining trend of contaminant levels or are below thresholds of concern for species or human health.
- By 2050, 95% of the samples gathered across Puget Sound habitats exhibit contaminant levels below thresholds of concern for species or human health and show no increasing trends.

The threshold of concern adopted for TPCB in English sole is 8 ng/g ww. This threshold was adopted from the Washington Department of Health screening values for assessing contaminant risks in Washington State seafood, based on subsistence (high-volume) seafood consumers (McBride, 2017). Successful remediation according to this recovery plan would be achieved when most fish sampled from TBIOS monitoring sites (when the 95th percentile, or right-hand whisker of the boxplot, at each location in Figure 8) is below the 8 ng/g ww threshold. As Figure 8 illustrates, only English sole from three locations (Hood Canal, Vendovi Is., and S. Strait of Georgia), all outside Puget Sound's most populated basins currently satisfy this criterion. Although TPCB concentrations in English sole from all CB/NT and other TBIOS monitoring locations exceeded this threshold, the mean TPCB concentration from the three background locations inside the main and south basins (12.8 ng/g ww – CIRA, Port Madison, and Anderson Island) was near to this threshold.

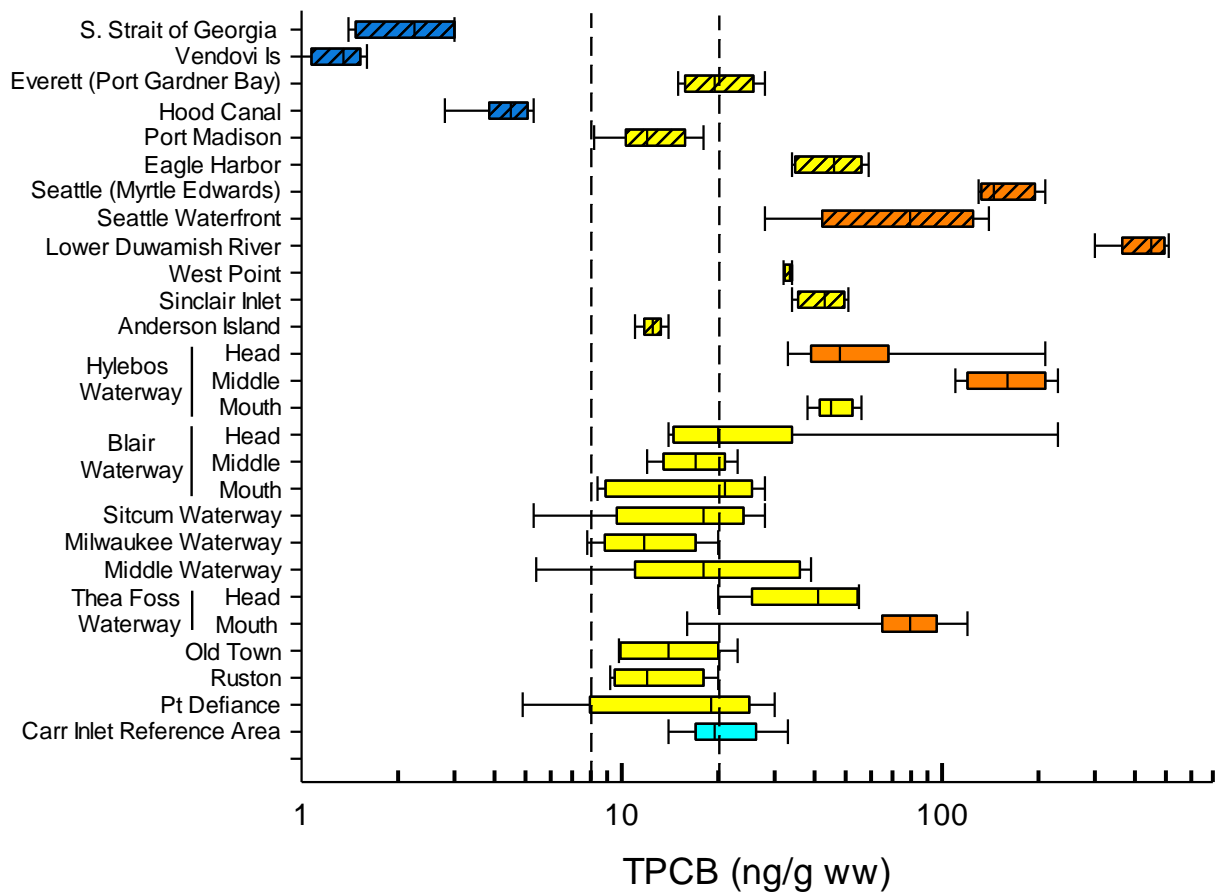


Figure 8. Comparison of TPCB in English sole from 15 CB/NT (boxplots with no-fills) and 12 TBiOS monitoring sites (hatched pattern) with the CIRA (cyan color). Boxplot whiskers indicate the 5th and 95th percentiles. Blue indicates locations exhibiting TPCB significantly below the CIRA; yellow indicates locations with TPCB concentration statistically indistinguishable from the CIRA, and orange indicates locations with TPCB significantly greater than the CIRA, based on an ANOVA of log-10-transformed TPCB by location, with a Holm-Sidak post hoc multiple comparison test using CIRA as the control. $\alpha=0.05$. Vertical dashed lines indicate the recovery target threshold for TPCB in English sole muscle tissue specified in the Toxics in Aquatic Life Vital Sign (8 ng/g; Puget Sound Partnership 2022b) and the mean CIRA TPCB concentration (21 ng/g).

A Thea Foss Focus Study

Investigating the trend of PCBs within the CB/NT in the years between 1984 and 2019 brings some perspective and context to understanding the effectiveness of remediation actions taken during that period. WDFW's TBiOS program has been monitoring PCBs in English sole from the mouth of the Thea Foss Waterway since 1991, and tissue samples have been collected, resected, and prepared using materially identical procedures and trawl locations with the 1984 Remedial Investigation. Combining Total Aroclor and TPCB data from CB/NT and TBiOS efforts provides a unique 38-year-long look at how PCBs have changed in fish living in that habitat (Figure 9). The Thea Foss mouth location was the most PCB-contaminated location measured in 1984, which originally provided impetus for its selection by WDFW for long-term TBiOS monitoring. WDFW initiated monitoring there in 1991 and it has continued since then.

Four sources of data were used to construct the Thea Foss PCB time trend illustrated in Figure 9.

- Original Thea Foss Total Aroclor values reported in the 1984 Remedial Investigation, shown as the dark blue triangle in 1984 (mean calculated from raw data; Tetra Tech Inc., 1985d),
- Aroclors from 1991-1997 (dark blue triangles) measured by the King County Environmental Laboratory (KCEL) for WDFW's TBIOS program (then called the Puget Sound Ambient Monitoring Program)
- PCB-congener data (cyan circles) measured by NOAA's NWFS Environmental Chemistry Program were compiled from:
 - 1997 through 2015 reported in West et al. (2017). A slight bias correction in congener-PCBs calculation from 1998-2001 related to a change in the congener methodology is described in West et al. (2017),
 - 2017, reported on the TBIOS Toxics in Aquatic Life Vital Sign (Puget Sound Partnership, 2022a)
 - 2019, measured in this study, and
 - 2021, (most recent monitoring data, WDFW unpublished)
- Aroclors measured by the EPA Manchester lab in 2019 (dark blue triangle).

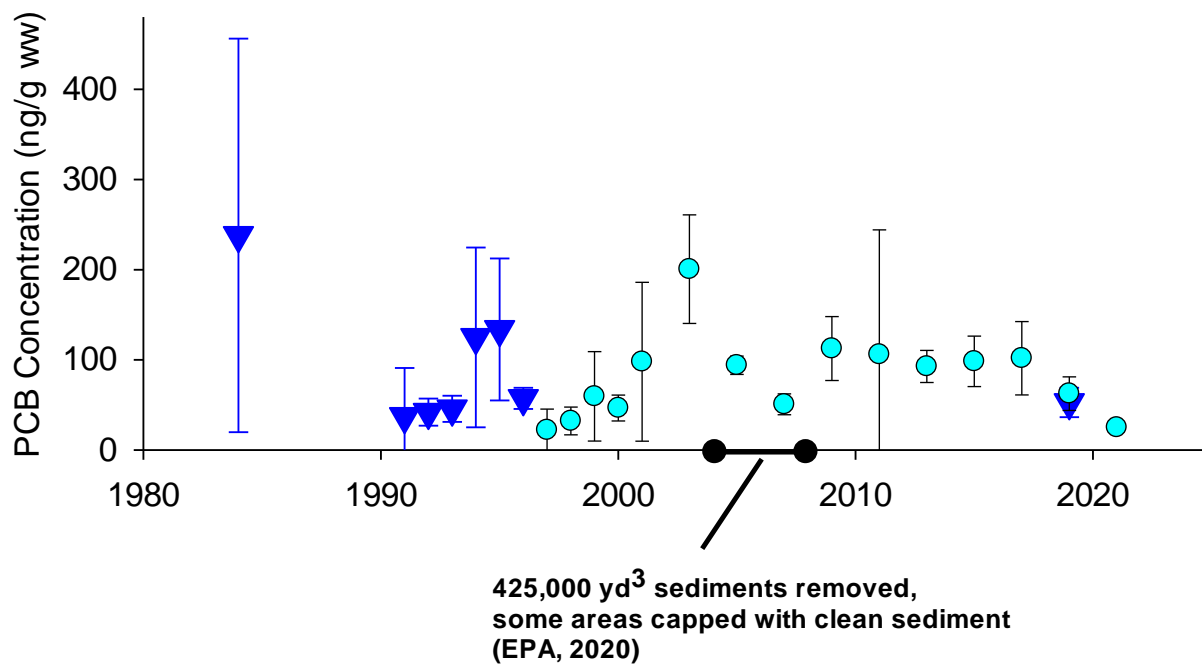


Figure 9. Time-Trend of PCBs in English sole sampled from the mouth of the Thea Foss waterway, from 1984-2019. PCBs were either measured by Aroclor quantitation (dark blue triangles) or congener methods (cyan circles) as detailed in the narratives above. A linear regression of PCBs through time was not significant ($p=0.26$).

The time trend plot in Figure 9 combines Total Aroclor data taken prior to 1997 (albeit with the Aroclor uncertainties described above) with TPCB data taken in 1997 and onwards, so caution is taken with the following interpretation. Two important characteristics of this plot are explored below: (1) a substantial drop in PCBs from 1984 to 1991, prior to any remediation activities in the Thea Foss or surrounding

areas, and (2) no significant trend in PCBs over 30 years, between 1991 and 2021 (linear regression of log-10 transformed PCB concentration by year; $p=0.26$).

This time trend plot indicates Total Aroclor concentrations in English sole from the mouth of Thea Foss Waterway dropped 6-fold from approximately 240 to 40 ng/g ww. in a seven-year period from 1984 to 1991. The primary characteristic of this pattern – a strong decline from the 1970s to a leveling off in the 1990s -- is consistent with PCB patterns reported for various biota worldwide, as well as in sediment cores in Puget Sound and San Francisco Bay. Stow et al (1994) reported exponential declines of PCBs in coho and Chinook salmon in Lake Michigan from 1974 to the late 1980s, with concentrations leveling off thereafter. Braune et al., (2005) and Bignert et al., (1998) reported similar declining patterns in biota from the Canadian Arctic and the Baltic Sea. PCBs have also declined significantly from the 1980s to the 1990s in bivalve mussels monitored in California (Melwani et al. 2014) and a site near Elliott Bay in central Puget Sound (Mearns 2002).

PCBs in other Puget Sound biota exhibited a similar large drop in PCBs from pre-1990s to the 1990s and leveling off thereafter. Stout and Beezhold (1981) reported PCBs in multiple species from Puget Sound fish collected in the mid-1970s that we can compare with more recent samples. English sole (80 fish analyzed individually) and coho salmon (10 individuals analyzed as a composite) muscle tissue from two locations matching later (1991-1992) WDFW TBIOS monitoring locations exhibited total PCB concentrations of 1,710 (Elliott Bay English sole) and 240 ng/g ww (central Puget Sound coho salmon). Lacking original data from Stout and Beezhold (1981) precludes a statistical comparison between the years, however English sole from WDFW's long-term PSEMP exhibited a mean total Aroclors concentration of 54 ng/g ww from Elliott Bay (6 composite samples each representing 20 fish from 1991 and 1992) and coho salmon from central Puget Sound exhibited 17 ng/g ww (6 composite samples each representing 5 fish from 1992). This comparison represents 32-fold and 14-fold reductions in PCBs in these species over a 17 year period in two disparate food webs (benthic and pelagic).

It is possible that measurement-bias related to changes in Aroclor methodologies during this period accounts for some of the declines just described. However, such declines in PCBs are also seen in sediment cores where concentrations are measured through time in a single analysis using a single, modern, congener approach. Lefkovitz et al. (1997) reported a PCB peak in sediments from three coring samples from the central basin of Puget Sound around 1960, with a sharp decline thereafter to the end of the core (approximately 1990). This pattern was also observed during this core-time period by Yee et al. (2006) in sediment cores from San Francisco Bay. Both authors primarily attribute the declines they observed from the 1960s to the 1990s to the federal ban on PCBs enacted in 1978, rather than regional or local sediment or other remediation efforts. Both studies employed congener PCB methods contemporaneous with the current study, with Lefkovitz et al. using GC-ECD, and Yee et al. using GC and high-resolution mass spectrometry. Congener analyses avoid problems associated with weathering of Aroclor patterns, and reconstructing PCB patterns through time in sediment cores avoids problems with changing bias through time related to changes in methods through time.

After the initial decline from 1984 to 1991, PCBs in English sole from the Thea Foss waterway mouth location leveled off. Remediation and source control for PCBs at Thea Foss began in 2002 through 2006, when 425,000 yds³ of sediments were removed, and some areas were capped with clean sands and gravels (USEPA 2020, Figures 2-5 to 2-7). For the 30-year period from 1991 through 2021, English sole from Thea Foss site exhibited no significant change in PCBs, including in the 14-year period after sediment dredging and capping had finished.

The lack of decline of PCBs in English sole from the mouth of the Thea Foss waterway is also apparent even if only the PCB congener methods are included (beginning in 1997). The congener methods

employed from 1997 through the present remove uncertainties associated with Aroclor methods. West et al (2017) reported an increasing trend of congener-PCBs at the Thea Foss waterway site (called “Tacoma City Waterway” in that document), using linear regression over the period from 1997 through 2015. The lower concentrations measured in Thea Foss English sole in the two most recent sampling years (2019 and 2021 -- shown in Figure 9) may indicate a declining trend or possibly variation observed similarly in other short timespans throughout the past 30 years.

Curiously, sediment remediation implemented in the Thea Foss Waterway in the mid-2000s appears not to have resulted in a decline in English sole PCBs over the ensuing 13 years, from 2008 to 2021. Two possible explanations seem viable: (a) the infilling and sediment capping, although spatially extensive, may not have remediated the full foraging area for these fish, and (b) English sole may be accumulating new PCB inputs to the system from terrestrial sources that may not be immediately discernable in sediments (using conventional sediment monitoring methods), but otherwise accumulating in their prey. English sole feed primarily on infaunal prey, especially including predatory polychaete worms and bivalve molluscs (Bizzaro et al. 2017). This is consistent with observations TBiOS staff have made by qualitatively examining English sole stomach contents from its monitoring stations over the past thirty years. Polychaete worms and the infaunal clam *Yoldia* sp. are commonly observed in English sole stomachs throughout Puget Sound. *Yoldia* spp. are shallow-burrowing clams, which can exhibit both deposit feeding and suspension feeding modes at the sediment-water interface (Davenport, 1988; Landry and Hickey, 1989). Both of these modes could increase their exposure to PCBs if newly settling allochthonous sediment particles (from terrestrial sources) are contaminated with PCBs. In effect, English sole prey such as *Yoldia* may be concentrating PCBs from this shallow, active particle-settling zone at a millimeter-sediment depth scale, whereas sediments sampled in the Thea Foss Waterway to document remediation integrate conditions throughout the top 2 to cm. PCB concentrations at the newly contaminated interface may be diluted with the cleaner sediment below in sediment grabs. Further investigations to evaluate the importance of PCB concentrations at the sediment-water interface, the organisms feeding there, and biomagnification of PCBs from these surface-sediment feeders to their predators are needed to clarify the significance of this pathway.

PCB Bioaccumulation in Additional Species

PCBs measured by TBiOS monitoring in CB/NT and nearby locations in three additional species (bay mussels (*Mytilus trossulus*), and two salmonids, juvenile Chinook salmon (*Oncorhynchus tshawytscha*), and cutthroat trout (*O. clarkii clarkii*)) corroborate and clarify the PCB distribution patterns in CB/NT revealed by English sole. Data from all three species point to PCB contamination throughout the Hylebos and Thea Foss Waterways, and support the lower PCB concentrations observed along the shoreline between the Blair and Middle Waterways and the shoreline from Old Town to Pt. Defiance.

Mussels

Caged, transplanted mussels were deployed according to TBiOS monitoring protocols (Langness et al., 2022) along CB/NT shorelines for two- to three-month periods from November to February throughout Puget Sound locations in the winters of 2012/13 (Lanksbury et al., 2014), 2015/16 (Lanksbury et al., 2017) and 2017/18 (Langness and West 2020). PCB data from these reports were compiled here to compare PCB concentrations in mussels among locations sampled within or near the CB/NT. Because mussels are only present in transplant locations for a short period, they do not represent the true contaminant concentrations wild mussels may exhibit from growing in these locations from the time they attached to their settlement substrate. However, because all transplanted mussels are derived

from the same aquaculture source (in Whidbey Basin) the initial baseline concentration of PCBs in all deployed mussels is the same, so differences observed in mussels retrieved after their winter exposure in transplant locations are attributable to accumulation from local PCB sources.

The initial PCB concentration in mussels deployed in these three surveys ranged from 1.0 to 4.6 ng/g ww, with a mean concentration of 2.5 ng/g ww. TPCB data for mussels deployed along CB/NT and nearby shorelines are illustrated in Figure 10. Each bubble represents an individual sample comprising a composite of 32 whole-body, shelled mussels. For the mussels retrieved from 52 CB/NT (or nearby) transplant locations in the three survey winters, 23% (n=12) of PCB concentrations were below 5 ng/g, indicating little or no net accumulation of PCBs relative to the initial condition during the exposure period. Fifty-two percent (n=27) of locations exhibited small increases, up to three-fold (15 ng/g ww) from the initial condition, and 25% of locations exhibited net accumulation from three- to seven-fold (Figure 10). This last group with the greatest net accumulation of PCBs had been deployed in the Hylebos Waterway, the shoreline near its mouth, and the mouth of Thea Foss Waterway.



Figure 10. TPCB concentration from caged/deployed mussels in CB/NT and nearby locations from WDFW's long-term mussel monitoring program. Samples years 2013, 2016, 2018 and 2020.

Salmonids

TBiOS sampled seaward-migrating juvenile Chinook salmon in 2013 (O'Neill et al., 2015) and 2016 (Puget Sound Partnership, 2022d), and cutthroat trout in 2013 (WDFW, unpublished data) in the CB/NT and nearby areas, as part of its long-term monitoring of toxics in Puget Sound salmonids. Monitoring persistent organic pollutants such as PCBs in Chinook salmon is particularly important because Chinook salmon originating from Puget Sound rivers, including the samples reported herein (likely originating from the Puyallup/White River) are listed as “threatened” under the US federal Endangered Species Act and they are recognized both as a pathway for persistent organic pollutants to, and a limiting prey for, ESA-endangered Southern Resident Killer Whales. Although most areas of Puget Sound are closed to retaining cutthroat trout, WDFW’s fishing rules allow retention of cutthroat from some areas of Puget Sound.

Five cutthroat trout were all taken from a beach seine near the Chinook Landing Marina, in the mouth of the Hylebos Waterway in 2013. Cutthroat ranged in size from 195 to 340 mm (fork length) indicating that at least four of these fish were likely greater than two years of saltwater age. PCBs were analyzed on individual whole-body cutthroat, using the TPCB analytical techniques described for English sole herein. TPCB concentration in these five samples ranged from 190 to 860 ng/g ww (mean of 530 ng/g ww; orange bar in Figure 11); the four largest of these individuals exhibited the greatest TPCB concentrations out of 1,296 whole-organism samples representing 20 species TBiOS has sampled for PCBs across Puget Sound from 1994-2018. The high concentrations in these fish most likely result from a combination of their residence (non-migratory behavior) in a contaminated habitat, high trophic level, and high lipid levels.

Unlike juvenile Chinook salmon which spend a short period of their life in the lower river/estuary habitat during their seaward migration, cutthroat trout tend to remain as residents in nearshore marine habitats, exhibiting high site fidelity (Losee et al., 2018). Although they exhibit anadromous spawning behavior, returning to their natal streams to spawn, they have been shown to return to a small geographic range in their saltwater, nearshore foraging habitat after spawning (Losee et al., 2018). The example of the cutthroat trout illustrates the risk to PCB exposure any organisms may experience if they reside in contaminated habitats such as the Hylebos Waterway.

PCB concentrations in 25 whole-body⁶ juvenile Chinook composites from the CB/NT and nearby locations ranged from 16 to 190 ng/g ww. The greatest concentration occurred in juvenile Chinook sampled from the same Hylebos Waterway location as the cutthroat trout. Three samples from that location ranged from 72 to 170 ng/g ww, with a mean of 110 ng/g ww (green bar, Figure 11). Although the small size (57-116 mm SL) of juvenile Chinook salmon sampled by TBiOS likely indicates these fish had only resided in the CB/NT area for a few weeks, their PCB concentrations (mean concentrations, 60, 160, and 30 ng/g ww for the Hylebos head, middle and mouth samples) were in a similar range to the English sole reported herein that have been accumulating PCBs for some years. Other juvenile Chinook salmon exhibiting intermediate (30-90 ng/g ww) concentrations were observed in the Thea Foss Waterway, Middle Waterway and nearby, in the Hylebos Waterway, and along the shoreline just outside and to the north of its mouth. Lowest (0-30 ng/g ww) TPCB concentrations in juvenile Chinook were observed near the Old Town location, near the former Milwaukee Waterway, and in Blair Waterway.

⁶ some had fragments of muscle, gill, brains, or other organs removed for other studies

Juvenile Chinook salmon were sampled during a life stage when they were actively migrating from their natal river (likely the Puyallup/White River or Hylebos Creek) through the CB/NT estuaries to the greater Puget Sound. Because of this we can be less certain their body burden reflected the specific location where they were collected. At the larger spatial scale of river systems however, O'Neill et al. (2015) and Carey et al. (2022) reported high TPCB concentrations in juvenile Chinook salmon from the Hylebos Waterway and shoreline areas around the mouth of the Puyallup River area in 2016 compared to 11 other river systems in Puget Sound (also reported on the Puget Sound Vital Sign, Puget Sound Partnership, 2022d). Juvenile Chinook salmon the Hylebos Waterway and the nearshore areas around the industrial waterways of CB/NT exhibited whole-body concentrations of TPCB statistically indistinguishable from juvenile Chinook sampled from the highly PCB-contaminated Lower Duwamish River Superfund Site. TPCBs in juvenile Chinook salmon from CB/NT areas are some of the highest concentrations observed by TBIOS monitoring from twelve major river systems sampled recently (2013-2018) in river mouth/estuary/nearshore habitats; the highest concentrations measured in these fish constitute levels high enough to kill them or impact their growth (Carey et al., 2022).

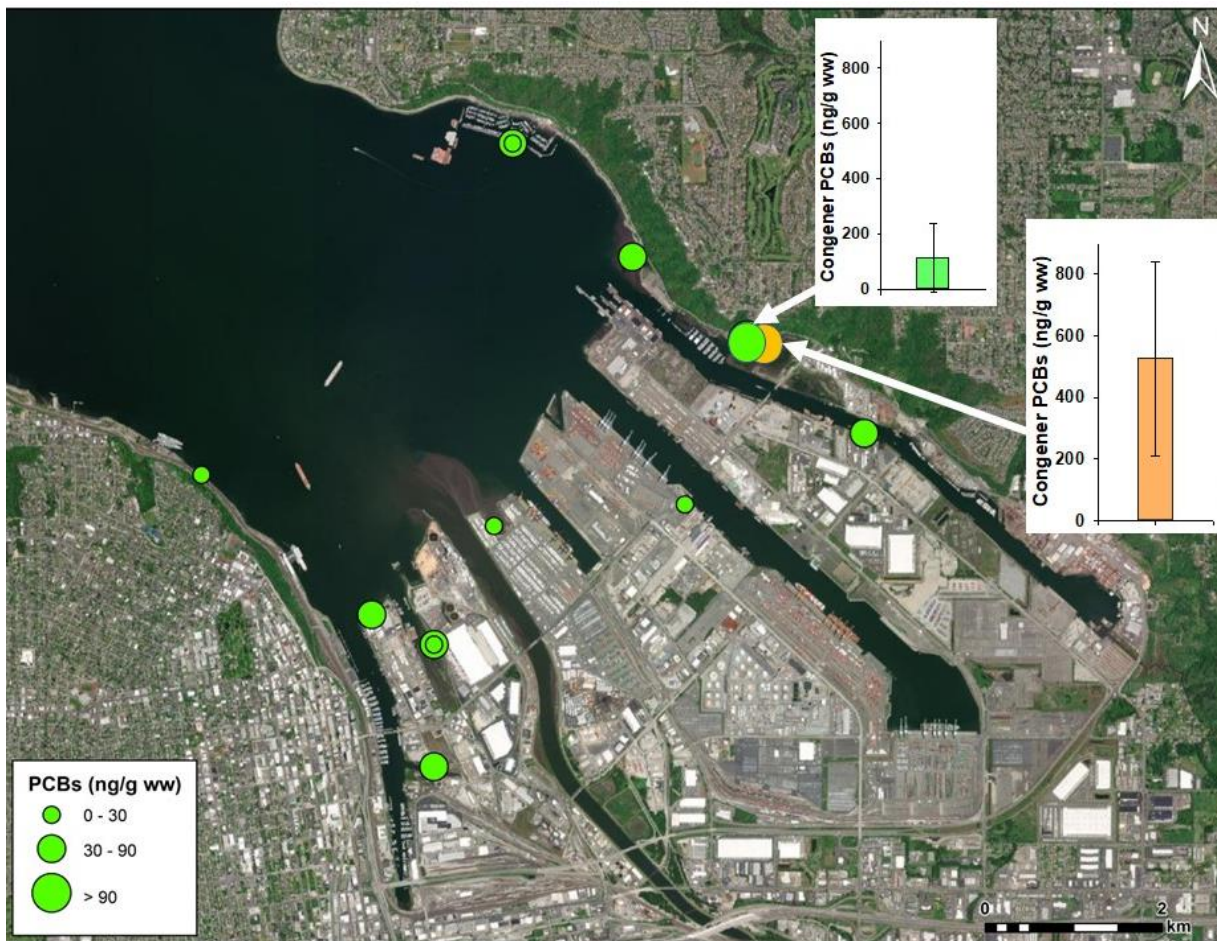


Figure 11. TPCB concentration in two salmonid species; cutthroat trout (orange bar/circle; *Oncorhynchus clarkii clarkii*) and juvenile Chinook salmon (green bar/circles; *O. tshawytscha*) sampled in 2013 and 2016. Bars indicate mean concentration \pm 95% confidence interval for locations with multiple samples.

Conclusions

Although the overall CB/NT recovery goal was met⁷, substantial areas of contamination in the CB/NT remain. Multiple lines of evidence presented here clearly indicate CB/NT's Operable Unit 1 continues to exhibit significant PCB contamination in fish tissue in two of its industrial waterways, Hylebos and Thea Foss. Further, there is no evidence to indicate PCB contamination has declined in English sole from the Thea Foss Waterway over the past 30 years, the only location for which long-term PCB monitoring data are available. This observation is despite significant sediment remediation efforts in that waterway. PCB contamination of English sole from Thea Foss may persist possibly because PCB contamination occurs in sediments available to fish but not successfully remediated, or fish have accumulated uncontrolled new inputs to the system from terrestrial sources. PCB contamination levels in English sole from both Hylebos and Thea Foss were similar to English sole from Puget Sound's other most highly contaminated areas, the Duwamish River, and Elliott Bay's shoreline along the Seattle Waterfront in 2019. These conclusions of continued, significant PCB contamination in the CB/NT are additionally supported by contemporaneous monitoring data in three other species, bay mussels, cutthroat trout, and juvenile Chinook salmon.

PCB contamination of English sole persists in the Blair Waterway, although the contamination status of that waterway is less clear. The number of samples exhibiting PCB concentrations above the CIRA (37%, including the high, 230 ng/g ww value) from the head of the Blair suggest English sole continued to accumulate PCBs from that area at concentrations that can greatly exceed the recovery goal.

Hylebos, Thea Foss, and Blair Waterways cover a substantial area of the industrialized waterways in Operable Unit 1 suggesting the contamination is a significant contributor to reduced habitat quality for aquatic species in the area. Cutthroat trout and juvenile Chinook salmon exhibited spatial patterns of PCB concentrations similar to English sole, indicating continued contamination in these CB/NT habitats, with reported levels in the cutthroat and juvenile Chinook from the Hylebos Waterway great enough to impair their health or kill them.

English sole in Sitcum Waterway, the shoreline areas fronting the mostly in-filled Milwaukee, Middle, and St. Paul Waterways, and the three shoreline areas along the western shoreline of Commencement Bay have met the objective of reaching PCB levels congruent with Puget Sound background conditions, as defined by PCB levels in the Carr Inlet Reference Area. Recovery of the Milwaukee/Middle/St. Paul shoreline likely resulted from the waterway infilling remedies implemented there. High PCB concentrations in English sole from the three western shoreline locations were not reported in the 1984 RI, and they appear to have remained low since that time.

These CB/NT locations meeting the CIRA target also exhibited PCB concentrations similar to two other reference or background locations monitored by TBiOS in the central and southern basins of Puget Sound, supporting a conclusion that these CB/NT locations currently exhibit ambient English sole PCB levels for these basins. English sole from none of the CB/NT locations or TBiOS locations in the central and southern Puget Sound basins have met a separate PCB recovery goal identified in the Puget Sound Partnership's Toxics in Aquatic Life Vital Sign.

⁷ the average site-wide PCB concentration from all English sole in the 15 sampling locations of Operable Unit 1 was statistically indistinguishable from the Carr Inlet Reference Area

Literature Cited

- Beliveau, A. F. (2001). PCB analyses needs for risk evaluations. *In*: Robertson, L.W. and L.G. Hanses (eds). PCBs: Recent Advances in Environmental Toxicology and Health Effects. Lexington, KY, USA, The University Press of Kentucky: 423-428.
- Bignert, A., et al. (1998). Temporal trends of organochlorines in Northern Europe, 1967-1995. Relation to global fractionation, leakage from sediments and international measures. *Environmental Pollution* **99**(2): 177-198.
- Butcher, J. B., et al. (1997). Use of historical PCB Aroclor measurements: Hudson River fish data. *Environmental Toxicology and Chemistry*. **16**(8): 1618-1623.
- Braune, B. M., et al. (2005). Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: An overview of spatial and temporal trends. *Science of The Total Environment* **351-352**: 4-56.
- Davenport, J. (1988) The feeding mechanism of *Yoldia (=Aeuiyoldia) eightsi* (Courthouy). Proceedings of the Royal Society of London. Series B., Biological Sciences, Vol. 232(1269): 431-442.
https://edisciplinas.usp.br/pluginfile.php/116119/mod_resource/content/1/youlida.pdf
- Carey, A., et al., (2022). Juvenile Chinook salmon accumulate harmful levels of toxic contaminants during their seaward migration to Puget Sound. Presentation to the 2022 Salish Sea Ecosystem Conference, 4/26/2022.
- Landry, M.R. and B.M. Hickey. (1989). Coastal Oceanography of Washington and Oregon. Elsevier Oceanography Series, Vol 47. Elsevier Science Publishers B.V. New York, NY. ISBN 0-444-87308-2.
- Langness and J. E. West (2020). Stormwater Action Monitoring 2017/18 Mussel Monitoring Survey Final Report. Washington Department of Fish and Wildlife; WDFW Report Number FPT 20-13. Olympia, Washington, USA. 90 pp.
- Langness, M. et al., (2022). Quality Assurance Project Plan: Status and trends monitoring of marine nearshore mussels in the Puget lowland ecoregion for Stormwater Action Monitoring (SAM) 2021-2025. Washington Department of Fish and Wildlife Publication FPT 22-02. Olympia, Washington, USA. 76 pp.
- Lanksbury, J., et al. (2014). Mussel watch pilot expansion 2012/2013: a study of toxic contaminants in blue mussels (*Mytilus trossulus*) from Puget Sound Washington, USA. Olympia, WA, Washington Department of Fish and Wildlife Report FPT 14-08. Olympia, Washington, USA. 55 pp.
- Lanksbury et al., (2017) Stormwater Action Monitoring 2015/16 Mussel Monitoring Survey: Final Report. Washington Department of Fish and Wildlife; WDFW Report Number FPT 17-06. Olympia, Washington, USA. 126 pp.
- Lauenstein, G. G. and A. Y. Cantillo, Eds. (1993). Sampling and analytical methods of the National Status and Trends Program National Benthic Surveillance and Mussel Watch Projects. 1984-1992. Silver Spring, MD, National Oceanic and Atmospheric Administration Report No. NOS ORCA 71. 187 pp.

- Lefkovitz, L. F., et al. (1997). Historical trends in the accumulation of chemicals in Puget Sound. NOAA Technical Memorandum NOS ORCA 111_NOAA, National Status and Trends Program. 159 pp.
- Lopez-Avila, V., et al. (1987). Single-laboratory evaluation of Method 8080 -- organochlorine pesticides and PCBs (polychlorinated biphenyls), Environmental Monitoring Systems Laboratory Office of Research and Development. US Environmental Protection Agency, Las Vegas, NV 89193: 252. Report No. EPA/600/4-87/022.
- Losee, J. P., et al. (2018). Size, age, growth and site fidelity of anadromous cutthroat trout *Oncorhynchus clarkii clarkii* in the Salish Sea. *Journal of Fish Biology* **93**(5): 978-987.
- McBride, D. (2017). Human health evaluation of contaminants in resident fish from the Hanford Reach of the Columbia River (Draft report). Olympia, WA, 98504, Washington State Department of Health. <https://doh.wa.gov/sites/default/files/legacy/Documents/Pubs//334-379.pdf>
- Mearns, A. (2002). Long-term Contaminant Trends and Patterns in Puget Sound, The Straits of Juan de Fuca and the Pacific Coast. Proceedings of the Puget Sound Research 2001 Conference. Paper #5A. 25 pp.
- Melwani, A. R., et al. (2014). Mussel watch update: Long-term trends in selected contaminants from coastal California, 1977–2010. *Marine Pollution Bulletin* **81**(2): 291-302.
- Muir, D. and E. Sverko (2006). Analytical methods for PCBs and organochlorine pesticides in environmental monitoring and surveillance: a critical appraisal. *Analytical and Bioanalytical Chemistry* **386**(4): 769-789.
- O'Neill, S. M., et al. (2015). Toxic contaminants in juvenile Chinook salmon (*Oncorhynchus tshawytscha*) migrating through estuary, nearshore and offshore habitats of Puget Sound. Department of Fish and Wildlife Report No. FPT 16-02 132. Olympia, Washington, USA. 132 pp.
- Puget Sound Partnership (2022a). Contaminants in English sole Vital Sign Indicator. Lead Reporter Louisa Harding, Washington Department of Fish and Wildlife, Olympia, WA, USA. Last updated May 23, 2022. <https://vitalsigns.pugetsoundinfo.wa.gov/VitalSignIndicator/Detail/48>.
- Puget Sound Partnership (2022b). Toxics in Aquatic Life Vital Sign. Reporter James E. West, Washington Department of Fish and Wildlife. Last updated May 3, 2022. <https://vitalsigns.pugetsoundinfo.wa.gov/VitalSign/Detail/28>
- Puget Sound Partnership (2022c). 2022-2026 Action Agenda: An update to the Puget Sound recovery plan for protected habitat, thriving species, and healthy people. <https://www.psp.wa.gov/2022AAupdate.php>
- Puget Sound Partnership (2022d). Contaminants in Juvenile Salmon Vital Sign Indicator. Lead Reporter Sandra M. O'Neill, Washington Department of Fish and Wildlife, Olympia, WA, USA. Last updated May 23, 2022. <https://vitalsigns.pugetsoundinfo.wa.gov/VitalSignIndicator/Detail/49>.

Sather, P. J., et al. (2001). Similarity of an Aroclor-Based and a Full Congener-Based Method in Determining Total PCBs and a Modeling Approach To Estimate Aroclor Speciation from Congener-Specific PCB Data. *Environmental Science and Technology* **35**: 4874-4880.

Sather, P. J., et al. (2003). Congener-Based Aroclor Quantification and Speciation Techniques: A Comparison of the Strengths, Weaknesses, and Proper Use of Two Alternative Approaches. *Environmental Science & Technology*. **37**(24): 5678-5686.

Stout, V. F. and F. L. Beezhold (1981). Chlorinated hydrocarbon levels in fishes and shellfishes of the northeastern Pacific Ocean, including the Hawaiian Islands. *Marine Fisheries Review* **43**(1): 1- 22.

Sloan, C. A., et al. (2014). Northwest Fisheries Science Center's Analyses of Tissue, Sediment, and Water Samples for Organic Contaminants by Gas Chromatography/Mass Spectrometry and Analyses of Tissue for Lipid Classes by Thin Layer Chromatography/ Flame Ionization Detection. NOAA Technical Memorandum NMFS-NWFSC-125. 81 pp.

Stow, C. A., et al. (1994). PCB concentration trends in Lake Michigan coho (*Oncorhynchus kisutch*) and chinook salmon (*O. tshawytscha*). *Canadian Journal of Fisheries and Aquatic Sciences* **50**: 1384-1390.

Tetra Tech. Inc. (1985a). Commencement Bay nearshore/tideflats remedial investigation. Volume 1. Olympia, WA, Prepared for Washington State Department of Ecology.

Tetra Tech. Inc. (1985b). Commencement Bay nearshore/tideflats remedial investigation. Volume 2. Olympia, WA, Prepared for Washington State Department of Ecology.

Tetra Tech. Inc. (1985c). Commencement Bay nearshore/tideflats remedial investigation. Volume 3. Olympia, WA, Prepared for Washington State Department of Ecology.

Tetra Tech. Inc. (1985d). Commencement Bay nearshore/tideflats remedial investigation. Volume 4. Olympia, WA, Prepared for Washington State Department of Ecology.

USEPA (1989). Superfund Record of Decision: Commencement Bay Nearshore/Tideflats, WA. Report No. EPA/ROD/R10-89/020. US Environmental Protection Agency
401 M Street, S.W., Washington, DC, 20460. 248 pp.

USEPA (2020). Fifth five-year review report for Commencement Bay Nearshore/Tideflats Superfund site, Pierce County, Washington, US Environmental Protection Agency, Region 10.

Washington Department of Fish and Wildlife (in prep) (a). Toxics Biological Observation System (TBIOS) for the Salish Sea: Standard Operation Procedure. Trawl Procedure for the Collection of Benthic and Demersal Fish and Macroinvertebrates. 21 pp.

Washington Department of Fish and Wildlife (in prep) (b). Toxics Biological Observation System (TBIOS) for the Salish Sea: Standard Operation Procedure. Collection of English Sole Tissue Samples for Analytical Chemistry. 21 pp.

West, J. E., et al. (2017). Time Trends of Persistent Organic Pollutants in Benthic and Pelagic Indicator Fishes from Puget Sound, Washington, USA. *Archives of Environmental Contamination and Toxicology* **73**(2): 207-229.

Wischkaemper, H. K., et al. (2013). US EPA Region 4 Technical Services Section Issue Paper for Polychlorinated Biphenyl Characterization at Region 4 Superfund and RCRA Sites, US EPA Region 4 Technical Services Section: 78. [U.S. EPA Region 4 Technical Services Section Issue Paper for Polychlorinated Biphenyl Characterization at Region 4 Superfund and RCRA Sites.](#)

Yee, D. (2011). Age estimates and pollutant concentrations of sediment cores from San Francisco Bay and wetlands. A Technical Report of the Regional Monitoring Program: SFEI Contribution 652. San Francisco Estuary Institute, Oakland, CA. 45pp + Appendices A, B and C.

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