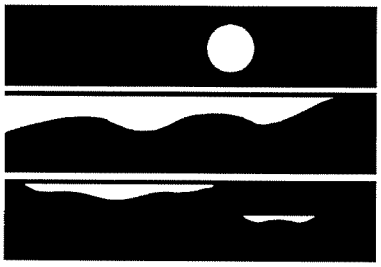

Washington State Pesticide Monitoring Program

Pesticides and PCBs in Marine Mussels, 1995

March 1996

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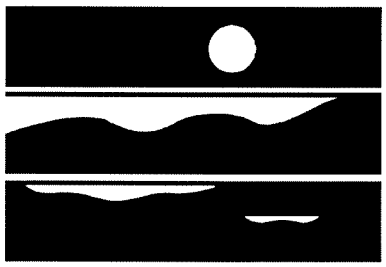

Washington State Pesticide Monitoring Program

Pesticides and PCBs in Marine Mussels, 1995

by
Art Johnson
and
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Olympia, Washington 98504-7710

March 1996

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Abstract

Forty-three pesticides or breakdown products and polychlorinated biphenyls (PCBs) were analyzed in mussels (*Mytilus trossulus*) collected from five sites in Puget Sound and one site at the mouth of the Columbia River. DDT, metabolites DDE and DDD, dieldrin, endosulfan, chlordane, gamma BHC (lindane), and PCBs were the most frequently detected.

The largest number of compounds and highest concentrations were found in Commencement Bay's Hylebos Waterway. The least contaminated site was Padilla Bay in north Puget Sound.

The results are compared to criteria for protection of human health and wildlife, and to data from historical and current monitoring programs. Some information is provided on the use, chemical behavior, and interrelationships of the compounds detected.

Summary

The Department of Ecology initiated the Washington State Pesticide Monitoring Program (WSPMP) in 1992 to characterize pesticide residues in surface water and ground water throughout the state. Sampling was extended to marine waters for the first time in 1995 with a collection of bay mussels (*Mytilus trossulus*; formerly *M. edulis*) from five sites in Puget Sound and one site at the mouth of the Columbia River. A composite sample of 30 or more mussels from each site was analyzed for 43 bioaccumulative pesticides or breakdown products and polychlorinated biphenyls (PCBs).

Twenty pesticide compounds and three PCB mixtures were detected. Mussels from all locations had detectable concentrations of DDT and/or metabolites DDE and DDD, and PCBs. Other frequently identified compounds were dieldrin, endosulfan, chlordane, and gamma BHC (lindane).

The largest number of compounds and highest concentrations were found in mussels from Commencement Bay's Hylebos Waterway. Data from other locales suggest the endosulfan levels in this waterway were unusually elevated. The least contaminated site was Padilla Bay in north Puget Sound. Other areas sampled included the West Duwamish Waterway in Elliott Bay, Chambers Creek mouth in south Tacoma, lower Budd Inlet at Olympia, and the Columbia River mouth at Ilwaco.

Currently-used pesticides detected in the mussel samples were the insecticides endosulfan and lindane, the fungicide pentachlorophenol (as the metabolite pentachloroanisole), and the herbicide DCPA (dacthal). The first three pesticides are among the most heavily used in the Puget Sound basin. As of 1989, Washington State had the highest annual rate of dacthal application in the U.S. PCBs and the remaining compounds are either banned or severely restricted in use, their presence being primarily due to long-term persistence.

Pesticide concentrations were well within U.S. Environmental Protection Agency (EPA) human health criteria for a 10^{-6} (one chance in a million) excess lifetime cancer risk. At all sites except Padilla Bay, total PCB concentrations exceeded the 10^{-6} risk level (6 - 70 ug/Kg vs. 1.4 ug/Kg; parts per billion). These data have been provided to the Washington State Department of Health, following routine WSPMP practice.

None of the mussel samples had pesticide or PCB residues that would be considered a concern for consumption by wildlife. NOAA has proposed a link between biological abnormalities they observe in Elliott Bay and Commencement Bay mussels and exposure to pesticide/PCB residues comparable to those detected in the present survey; polyaromatic hydrocarbons were also implicated.

Historical data show there has been a substantial decrease in PCB levels in Duwamish Waterway/Elliott Bay mussels and other media, as well as a general decline in PCB and DDT compounds for the greater Puget Sound. Dacthal -- only detected in mussels from Ilwaco -- occurs in high concentrations in upper Columbia River fish but appears to be a minor contaminant in the lower river.

Results from this limited survey show a higher detection frequency of pesticides in mussels than in the clams, fish, and sediment samples analyzed as part of the Puget Sound Ambient Monitoring Program (PSAMP). In the case of fish and sediment this may be largely due to differences in sampling location.

Recommendations

1. Include mussels in future WSPMP sample collections as a marine indicator for pesticides and PCBs.

2. Re-sample mussels from Hylebos Waterway to verify elevated pesticide and PCB concentrations.

3. Add Chambers Creek (WA-12-1010) and the lower Columbia River (WA-CR-1010) to the 1996 water quality limited list (303d) for exceeding the human health edible tissue criterion for PCBs.

Acknowledgements

Dave Serdar assisted in collecting the mussel samples for this report. Arrangements for contract laboratory services were made by Stuart Magoon of Manchester Laboratory.

This report benefitted from review by Ken Dzinbal, Dale Norton, and Larry Goldstein.

Final formatting and proofreading were done by Joan LeTourneau.

Introduction

The Washington State Pesticide Monitoring Program (WSPMP) was begun by the Department of Ecology Environmental Investigations and Laboratory Services Program in 1992. The WSPMP goal is to characterize pesticide residues geographically and over time in surface water and ground water throughout Washington. Specific objectives are as follows:

- Identify and prioritize aquifers, lakes, and streams with known or potential pesticide contamination
- Quantify pesticide concentrations in high priority areas
- Document temporal trends in pesticide concentrations at selected sites
- Provide data to the Washington State Department of Health for assessment of potential adverse effects on human health
- Assess the potential for adverse effects on aquatic biota
- Construct and maintain a pesticide database for ground water and surface water in Washington State
- Provide information for the improvement of pesticide management in Washington

Surface water sampling has been focused on rivers, lakes, and streams near areas where pesticides are used. Water, fish tissue, and bottom sediments have been analyzed for up to 162 pesticides and toxic breakdown products (Davis, 1993; Davis and Johnson, 1994a&b; Davis et al., 1995).

In 1995, sampling was extended to marine waters with a small collection of bay mussels from Puget Sound and the Columbia River estuary. Mussels were selected for analysis because of their usefulness in other contaminant monitoring programs (e.g., Farrington et al., 1983; Rasmussen, 1995) and occurrence close to points of pesticide inputs from rivers and streams. Sampling was done during May to coincide with the period when pesticides are most frequently detected in Puget Sound tributaries (Davis and Johnson, 1994b).

Samples were obtained at six sites located near potential agricultural, urban, and/or industrial sources of pesticides (Figure 1):

- Padilla Bay, North Puget Sound
- West Duwamish Waterway, Elliott Bay, Seattle
- Head of Hylebos Waterway, Commencement Bay, Tacoma
- Mouth of Chambers Creek, South Tacoma
- Lower Budd Inlet, Olympia
- Mouth of Columbia River, Ilwaco

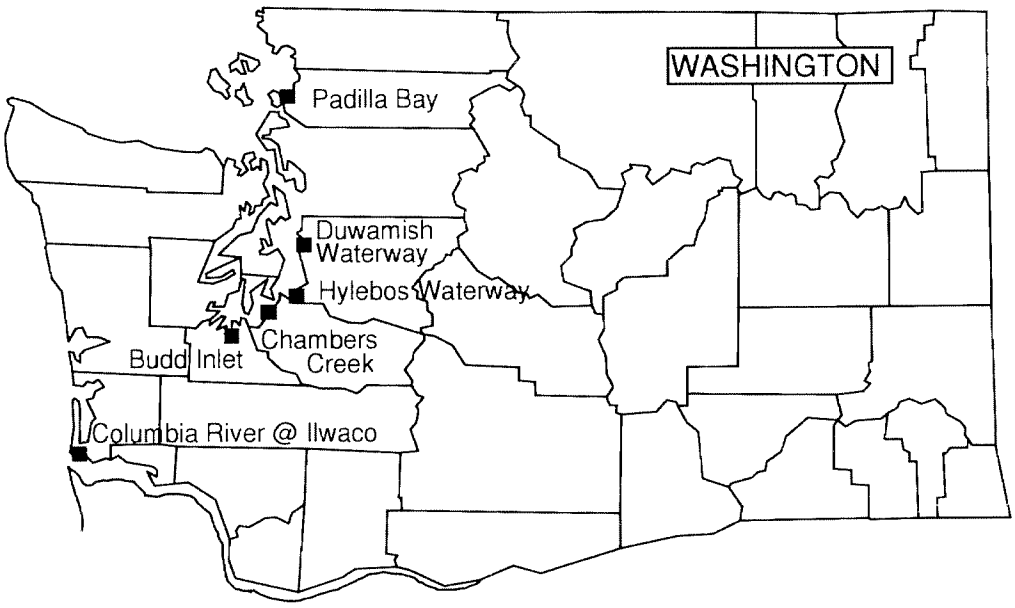

Figure 1. Location of 1995 WSPMP Mussel Samples

For each site, the entire soft parts were composited from 30 or more individual mussels. The composites were analyzed for 43 pesticide compounds that have been shown to be moderately-to-highly bioaccumulative (Table 1). Data were also obtained on polychlorinated biphenyls (PCBs). Appendix A has detailed information on sampling locations and sample size.

Methods of Sampling and Analysis

Mussels (*Mytilus trossulus*; formerly *M. edulis*) were scraped from rocks, pilings, or other structures. The samples were wrapped in aluminum foil, placed in plastic bags, and kept on ice. On return to the laboratory, the shells were washed with tap water to remove sediment and other debris, then rinsed with deionized water.

The soft parts from the largest individuals were removed with stainless steel scalpels, homogenized with a Kitchen-Aid food mixer, and poured into 8-oz. priority pollutant cleaned glass jars with teflon lid-liners. The samples were then frozen pending chemical analysis. Sample preparation equipment was washed with Liquinox detergent, then rinsed with tap water, deionized water, and pesticide-grade acetone.

The samples were analyzed by the California Department of Fish & Game, Water Pollution Control Laboratory. Tissue aliquots of 20-25 grams were extracted with acetone and cleaned up using Florisil columns and gel permeation chromatography. The 0%, 6%, and 50% Florisil fractions were analyzed separately by dual, dissimilar, capillary column gas chromatography with electron capture detection and all positives confirmed by gas chromatography/mass spectroscopy. Details of the method can be found in Rasmussen and Blethrow (1991) and Magoon (1993). Percent total lipids and percent non-polar lipids were determined according to EPA (1980) and Schneider (1992), respectively.

Quality of the Data

Karin Feddersen of the Ecology/EPA Manchester Laboratory reviewed the quality of the data. The review included adherence to sample holding times, results on method blanks, instrument tuning and calibration, precision data, and recovery of surrogate compounds and matrix spikes.

Table 1. Pesticides and PCBs Analyzed in 1995 WSPMP Mussel Samples

Compound	Quantitation Limit (ug/Kg (ppb))	Compound	Quantitation Limit (ug/Kg (ppb))
DDT & Analogs		Benzene Hexachloride	
4,4'-DDT	0.80	alpha BHC	0.24
4,4'-DDE*	0.60	beta BHC	0.80
4,4'-DDD**	1.5	delta BHC	0.50
4,4'-DDMU*	1.5	gamma BHC	0.40
2,4'-DDT	0.65		
2,4'-DDE*	1.0	Misc. Chlorinated Pesticides	
2,4'-DDD*	1.5	hexachlorobenzene	0.15
dichlorobenzophenone*	20	DCPA (dacthal)	0.25
dicofol	6.5	oxadiazon	0.30
methoxychlor	3.0	tetradifon	0.20
		mirex	0.80
Cyclodienes		toxaphene	0.15
aldrin	0.30		
dieldrin	0.15	Organophosphates	
endrin	0.10	diazinon	3.0
endrin aldehyde*	0.15	chlorpyrifos	1.5
endrin ketone*	0.60	ethion	3.5
endosulfan I	0.18	ethylparathion	0.45
endosulfan II	0.65	methylparathion	0.30
endosulfan sulfate*	1.5		
cis-chlordane	0.50	Phenols	
trans-chlordane	0.50	pentachloroanisole***	0.55
cis-chlordene	0.30		
trans-chlordene	0.35	Polychlorinated Biphenyls	
cis-nonachlor	0.75	PCB-1016	6.0
trans-nonachlor	0.30	PCB-1221	6.0
oxychlordane*	0.50	PCB-1232	6.0
heptachlor	0.30	PCB-1242	6.0
heptachlor epoxide*	0.30	PCB-1248	6.0
		PCB-1254	6.0
		PCB-1260	6.0

* breakdown product

** breakdown product and insecticide

*** metabolite of pentachlorophenol

Three problems were identified through the review. Oxychlordane was detected in both the method blank and field samples. Because concentrations in the samples were less than five times the blank, oxychlordane is reported as being not detected. Matrix spike recoveries of endrin aldehyde, ethylparathion, and ethion were low. Failure to detect these compounds in the samples may reflect a shortcoming in the analysis. Finally, some results were qualified as estimates if the relative percent difference between dual capillary columns was greater than 30%. The data review and complete pesticide, PCB, and lipid data are in Appendix B.

The potential contribution of analytical variability to the precision of the data reported here can be estimated from results on a split (duplicate) sample (Table 2). With the exception of cis-nonachlor, agreement between duplicate analyses was within 20% or better, showing good precision. Results that differ by 20% or less may be due to laboratory variability rather than differences in the level of contamination between sampling sites.

Table 2. Precision of the Pesticide/PCB Data* (ug/Kg (ppb) wet wt.)

Compound	Analysis #1		Analysis #2		RPD**	Compound	Analysis #1		Analysis #2		RPD**
4,4'-DDT	5.5	NJ	5.7	NJ	4%	alpha BHC	0.10	J	0.12	J	18%
4,4'-DDE	5.9		6.4		8%	gamma BHC	0.08	J	0.09	J	12%
4,4'-DDD	3.7		3.8		3%						
2,4'-DDT	1.4		1.7		19%	hexachlorobenzene	0.40		0.50		4%
2,4'-DDE	0.14	J	0.15	J	7%						
						pentachloroanisole	0.28	J	0.31	J	10%
dieldrin	0.71		0.77		8%						
endosulfan I	22		25		13%	PCB-1248	18		15		6%
endosulfan II	12		13		8%	PCB-1254	42		50		17%
endosulfan sulfate	7.4		8.4		13%	PCB-1260	6	J	5	J	18%
cis-chlordane	1.2		1.2		0%						
trans-chlordane	1.0		1.0		0%	% lipid	1.3		1.1		17%
cis-nonachlor	0.26	J	0.37	J	35%	% non-polar lipid	0.41		0.36		13%
trans-nonachlor	0.99	NJ	1.1	NJ	11%	% moisture	85		86		1%

* Hylebos Waterway mussel sample no. 20-8043

** Relative Percent Difference = (range/mean) x 100

J = estimated value

N = tentatively identified

Results and Discussion

Pesticides and PCBs Detected

Results of the chemical analyses are summarized in Table 3. Twenty pesticides or breakdown products, and three PCB mixtures, were detected in the mussels. The PCB data are reported in terms of equivalent concentrations of commercial PCB mixtures, also known as Aroclors. The first two numbers (e.g., PCB-1254) denote the 12 carbons in biphenyl and the last two the percentage of chlorine by weight.

DDT and/or metabolites DDE and DDD, and PCBs were detected at all six sampling sites (Figure 2). Other frequently detected compounds (four-to-five sites) were dieldrin, endosulfan I, cis- and trans-isomers of chlordane, trans-nonachlor, and alpha BHC.

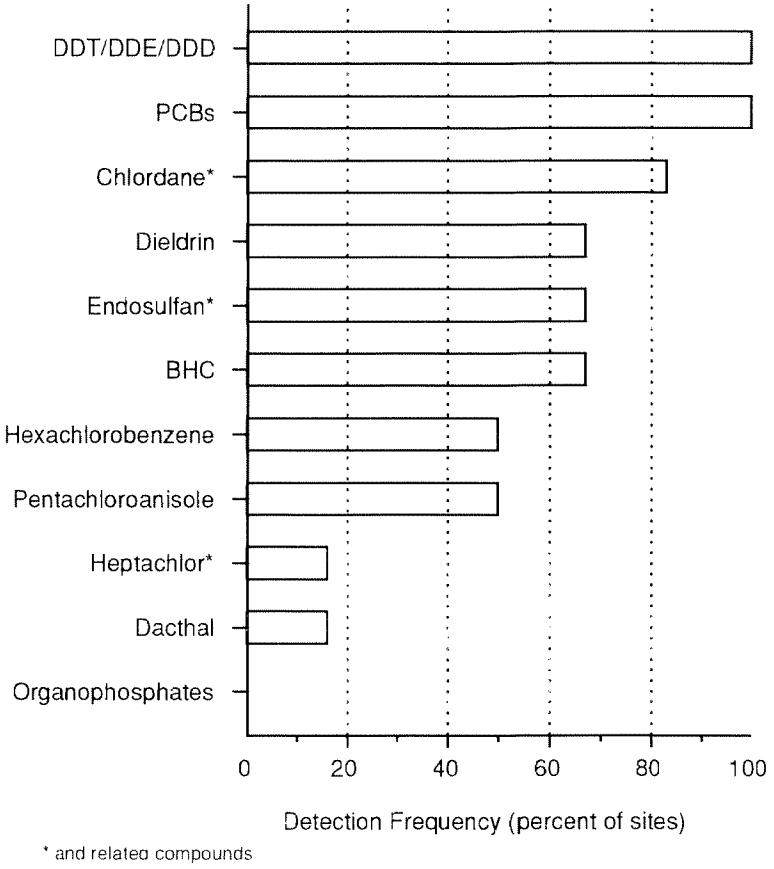

Figure 2. Detection Frequency of Pesticides/PCBs in Mussels

Table 3. Pesticides and PCBs Detected in 1995 WSPMP Mussel Samples (ug/Kg (ppb) wet wt.)

Compound	Padilla Bay	Duwamish Waterway	Hylebos Waterway	Chambers Creek	Budd Inlet	Ilwaco
DDT & Analogs						
4,4'-DDT	0.12 J	2.8 NJ	5.6 NJ	nd	nd	0.91
4,4'-DDE	0.57 J	1.4	6.2	0.33 J	0.70	6.5
4,4'-DDD	0.05 J	0.88 J	3.8	nd	0.45 J	1.8
2,4'-DDT	nd	0.61 J	1.6	nd	nd	0.25 J
2,4'-DDE	nd	nd	0.14 J	nd	nd	nd
Cyclodienes						
dieldrin	nd	0.22	0.74	0.16 N	nd	0.43 J
endosulfan I	0.15 J	nd	24	nd	0.66 NJ	0.18
endosulfan II	nd	nd	12	nd	nd	nd
endosulfan sulfate	nd	nd	7.9	nd	nd	nd
cis-chlordane	nd	0.97	1.2	0.37 J	0.49 J	0.33 J
trans-chlordane	nd	0.91	1.0	nd	0.58	0.50
cis-nonachlor	nd	0.20 J	0.32 J	nd	nd	nd
trans-nonachlor	nd	0.66 NJ	1.0 NJ	0.38	0.34	nd
heptachlor	nd	nd	nd	nd	nd	0.01 J
heptachlor expoxide	nd	nd	nd	nd	nd	0.14 J
Benzene Hexachloride						
alpha BHC	0.06 J	0.06 J	0.11 J	nd	0.07 J	nd
gamma BHC	nd	nd	0.08 J	nd	nd	nd
Misc. Chlorinated Pesticides						
hexachlorobenzene	0.03 J	nd	0.45	nd	nd	0.14 J
DCPA (dacthal)	nd	nd	nd	nd	nd	0.33
Phenols						
pentachloroanisole	nd	0.05 J	0.30 J	nd	0.15 J	nd
Polychlorinated Biphenyls						
PCB-1248	nd	nd	18	nd	nd	nd
PCB-1254	2 J	32	46	6	21	6 N
PCB-1260	nd	12 J	6 J	2 J	nd	nd
% lipid	0.8	1.1	1.2	1.0	1.4	1.1
% non-polar lipid	0.1	0.1	0.4	0.1	0.1	0.1
% moisture	90	87	86	87	84	89

nd = not detected J = estimated value N = tentatively identified

Heptachlor and DCPA (dacthal) were only detected in mussels from the mouth of the Columbia River. Isomers of the above compounds (endosulfan II, gamma BHC, and cis-nonachlor) or degradation products (endosulfan sulfate, heptachlor epoxide, and pentachloroanisole) were also occasionally identified.

None of the samples contained detectable levels of organophosphate (OP) pesticides. OPs are less persistent than the other compounds analyzed and have relatively low bioaccumulation potential. For example, diazinon, the most frequently detected OP in tributaries to Puget Sound (Davis and Johnson, 1994b), has bioaccumulation factors reported in the range of 5 to 150, compared to greater than 10,000 for PCBs and most chlorinated pesticides (CCREM, 1987; Tetra Tech, 1988). The biological half-life of diazinon in invertebrates and fishes can be less than one day (CCREM, 1987). OP pesticides are also infrequently detected (< 10% of samples) in Washington freshwater fish (Davis et al., 1995).

The largest number of pesticide compounds (17) and highest concentrations were found in mussels from Hylebos Waterway. The least contaminated site was Padilla Bay (6 compounds detected). Concentrations of DDT, endosulfan, chlordane, and related compounds exceeded 1 ug/Kg (part per billion) in the Hylebos sample. Total endosulfan concentrations were relatively high at 44 ug/Kg. Pesticides detected elsewhere were generally at concentrations less than 1 ug/Kg.

Hylebos mussels also had the highest level of total PCBs, 72 ug/Kg. Concentrations at other sites ranged from 2 - 54 ug/Kg total PCBs with the lowest concentration again occurring at Padilla Bay. PCB-1254 was the predominant mixture identified in all areas.

The elevated concentrations in mussels from Hylebos Waterway may be partly explained by the higher percentage of non-polar lipids compared to other samples (0.4% vs. 0.1%; Table 3). Non-polar organochlorine compounds have been shown to be primarily associated with this fraction of the total lipid pool (Schneider, 1982; Kawai et al., 1988).

Currently-Used Compounds*

Mussels showed the presence of four pesticides that are currently used: endosulfan, lindane, dacthal, and pentachlorophenol. Endosulfan is an insecticide/acaricide applied to fruit and deciduous trees, ornamentals, vegetables, and other crops. Endosulfan breaks down to endosulfan sulfate which is more persistent than the parent compound and comparably toxic. Lindane, also an insecticide, is comprised of 99% gamma BHC (benzenehexachloride). The alpha BHC isomer was banned from the formulation in 1977. Lindane has uses on the above-mentioned crops and prominent use in treating seed.

Dacthal is a selective herbicide for controlling broadleaf weeds and grasses. It is unusual among modern herbicides in being accumulated by aquatic organisms.

The primary metabolic product of the fungicide pentachlorophenol is pentachloroanisole. In 1984, the use of pentachlorophenol was restricted to preserving wood. Pentachloroanisole is relatively short-lived.

As of 1988, pentachlorophenol, endosulfan, and lindane were among the most heavily used pesticides in the Puget Sound basin (Tetra Tech, 1988). A national survey of herbicide usage in 1987-89 found Washington State had the highest annual rate of dacthal application (Gianessi and Puffer, 1991). Dacthal was only detected in mussels from the Columbia River estuary. Dacthal is used on a number of crops grown in eastern Washington, including onions, potatoes, and seed crops. Data on the levels of dacthal in the Columbia drainage are discussed later in this report.

Banned or Restricted Compounds*

PCBs and the majority of pesticides detected in the mussel samples are either banned or their use severely restricted. Their presence primarily reflects long-term persistence rather than recent application.

DDT was banned in 1972 after more than 20 years of extensive use on a broad spectrum of insects. Its most frequently detected metabolite, DDE, breaks down slowly, if at all, in aquatic environments. DDD, the other commonly reported degradation product, was also sold as the insecticide Rothane.

* This information taken from Callahan et al. (1979), CCREM (1987), EPA (1992), Schmitt et al. (1982), and Seyler (1994), except as noted.

Restrictions were placed on the uses of dieldrin, heptachlor, chlordane, and hexachlorobenzene in the 1970s. All use ended in the early 1980s, except for some continued use of heptachlor and chlordane for termites and fire ants. Technical chlordane is a complex mixture that includes cis- and trans- isomers of chlordane, chlordene, and nonachlor, heptachlor, and other closely related compounds. Oxychlordane and heptachlor epoxide are the primary degradation products.

Although the fungicide hexachlorobenzene is no longer used, there are some minor present-day sources. It is an unintended byproduct from production of other chlorinated compounds like carbon tetrachloride, tetrachloroethene, and trichloroethene. Hexachlorobenzene has also been reported as a trace impurity or breakdown product of some pesticides, including dacthal, pentachlorophenol, and lindane.

PCBs were widely used as insulating fluids, plasticizers, in inks and carbonless paper, and as heat transfer and hydraulic fluids. In 1979, EPA banned PCB manufacture, processing, and distribution, but allowed continued use in closed electrical systems. Regulations in 1982 and 1985 phased out use in electrical components, although these continue to be sources of PCBs to the environment from old, leaking equipment.

Human Health and Wildlife Criteria

Table 4 compares the pesticide and PCB concentrations found in mussels to criteria for protection of human health and wildlife from consumption of contaminated fish or shellfish. The human health criteria shown are the EPA 10^{-6} (one chance in a million) excess individual lifetime cancer risk values.* These are used in the 303(d) assessment of water quality limited waterbodies which Ecology updates every two years as required by the Clean Water Act. EPA health criteria are lacking for dacthal.

There are no adopted state or national numeric criteria for protection of fish and wildlife from adverse effects of pesticide or PCB residues in biota. The fish flesh criteria listed in Table 4 were developed by the state of New York to protect populations of fish-eating wildlife (Newell et al., 1987). The criteria shown are the lower of two values proposed, one for cancer risk, the other for non-carcinogenic effects. A cancer risk level of 10^{-2} was considered sufficient to prevent reductions in wildlife populations. Criteria for non-carcinogenic effects were calculated as no-observed-effect levels. The methods used to derive the New York criteria were recently selected to develop tissue residue guidelines in Canada (Environment Canada, 1994-draft)

*See EPA (1995) for a discussion of cancer risk assessment.

With one exception for dieldrin, the concentrations of pesticides and breakdown products detected in Puget Sound and Columbia River mussels are well within EPA human health criteria. The dieldrin concentration at Hylebos Waterway, 0.74 ug/Kg, exceeds the criterion of 0.65 ug/Kg, but only slightly.

Table 4. Mussel Results Compared to Human Health and Wildlife Criteria (ug/Kg (ppb) wet wt.)

Compound	Concentrations in Mussels	EPA Human Health Criteria	Fish-eating Wildlife Criteria
dieldrin	nd - 0.74	0.65	22
total PCBs	2 - 70	1.4	110
heptachlor	nd - 0.01	2.4	200
heptachlor epoxide	nd - 0.44	1.2	(hetachlor + epoxide)
alpha BHC	nd - 0.11	1.7	100
gamma BHC	nd - 0.08	8.2	(total BHC)
hexachlorobenzene	nd - 0.44	6.7	200
total chlordane*	nd - 3.5	8.3	370
total DDT**	0.33 - 17	32	200
endosulfan I/II	nd - 24	540	--
endosulfan sulfate	nd - 7.9	540	--
dacthal	nd - 0.33	- -	--
pentachloroanisole	nd - 0.30	90.2***	--

* sum of chlordane and nonachlor isomers
** DDT+DDE+DDD
*** pentachlorophenol

Mussels from all sites had total PCB concentrations that exceeded the human health criterion of 1.4 ug/Kg. Three locations -- Hylebos Waterway, Duwamish Waterway, and Budd Inlet -- had concentrations exceeding the 10^{-5} risk level of 14 ug/Kg. These waterbodies are on the 1994 '303(d) list for exceeding sediment PCB criteria. The PCB concentrations in Chambers Creek and Ilwaco mussels meet requirements for listing under '303(d). Given the precision of the PCB analysis, the 0.6 ug/Kg elevation above the criterion in Padilla Bay mussels is not significant. These data have been provided to the Washington State Department of Health, following routine WSPMP practice.

The criteria to protect wildlife populations are several orders of magnitude less restrictive than for individual human health. None of the mussel samples had concentrations approaching the wildlife criteria. Although values are lacking for endosulfan, dacthal, and pentachloroanisole, these compounds are relatively low in toxicity.

Effects on Mussels

NOAA reports that mussels from Elliott Bay and Commencement Bay have "impaired growth, reduced fecundity, and altered age-structure patterns" compared to mussels from non-urban bays (Krishnakumar et al., 1994; Kagley et al., 1995). NOAA's analysis of the tissues showed concentrations of total PCBs (38-64 ug/Kg), total DDT (2-4 ug/Kg) and "total pesticides"(2-4 ug/Kg; sum of aldrin, chlordane, dieldrin, heptachlor, and lindane) similar to present survey findings for these two sites*. Elevated levels of polyaromatic hydrocarbons were also reported. The authors concluded their "findings indicated potential linkages between exposure of mussels to chemical contaminants and observed biological effects". It remains to be demonstrated whether the chemicals analyzed were the cause of these effects or if other chemicals or factors are involved.

Historical Data

Four surveys report historical levels of pesticides and PCBs in mussels collected in or near the Duwamish Waterway, Hylebos Waterway, Budd Inlet, and the Columbia River mouth (Appendix C). Mowrer et al., (1977) surveyed PCBs in Puget Sound mussels in 1975. National Mussel Watch Programs analyzing chlorinated pesticides, PCBs, and a range of other contaminants were carried out in 1976-78 by the Scripps Institute of Oceanography and in 1986-88 by NOAA (Goldberg, 1978; Farrington, 1982,83; NOAA, 1989).

* NOAA data converted from dry wt. to wet wt. assuming 87% moisture (Table 2).

Ecology monitored chlorinated pesticides, PCBs, and metals in mussels from several locations in Puget Sound during 1979-82 (Hopkins et al., 1985).

Data from these programs may not be directly comparable to present survey results, because of differences in sampling sites, season of collection, and analytical methods. The older data generally indicate that levels of DDT, dieldrin, chlordane, hexachlorobenzene, and PCBs in Puget Sound and Columbia River mussels were, at most, only moderately higher during the 1970s and 80s. However, PCB concentrations in Duwamish Waterway/Elliott Bay mussels have undergone a substantial and consistent decrease since first sampled in 1975 (Figure 3). A number of other studies have shown that PCB levels have declined in Elliott Bay (Dexter et al., 1981; Hart Crowser, 1990; Matta et al., 1986; Mearns et al., 1988). A general downward trend has been observed in concentrations of PCBs and DDT compounds in the bottom sediments and marine mammals of Puget Sound (Lefkovitz et al., 1995; Calambokidis, 1995).

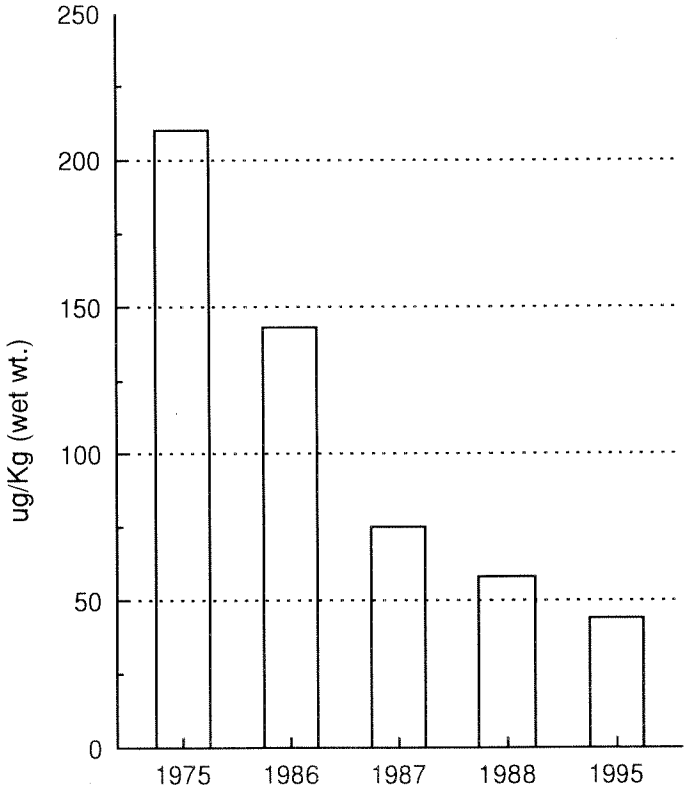

Figure 3. Total PCBs in Duwamish WW/Elliott Bay Mussels, 1975 - 1995 (see Appendix C for data sources)

Mussel watch programs also provide a perspective on what concentrations have occurred near significant sources of contamination. DDE concentrations have exceeded 100 ug/Kg in mussels near major California wastewater discharges and harbors, compared to 6 ug/Kg or less in the present survey (Martin, 1985). Up to 2,210 ug/Kg total PCBs have been reported in mussels from the highly contaminated New Bedford Harbor area of Buzzards Bay, Mass., vs. the 2 - 70 ug/Kg reported here (Farrington, 1983). NOAA calculated national mean values of 3 ug/Kg total DDT and 15 ug/Kg total PCBs from 145 sites in their 1986 Mussel Watch survey (Mearns et al., 1988).

One instance where substantial pesticide contamination may be indicated is endosulfan in Hylebos Waterway. Extensive monitoring results from California show this compound is infrequently detected in mussels. Moss Landing was characterized as an endosulfan "hotspot" when unusually high concentrations of 34 - 100 ug/Kg endosulfan I were found; agricultural sources were suspected (Martin, 1985). This level of contamination was approached by the endosulfan I concentration of 24 ug/Kg (44 ug/Kg total endosulfan) in Hylebos mussels.

Results from Other Monitoring Programs

Puget Sound

Contaminant monitoring in Puget Sound is done primarily by several agencies through the Puget Sound Ambient Monitoring Program (PSAMP). Since 1989, PSAMP has collected data on pesticides, PCBs, and other chemicals in littleneck clams (Dept. of Health), several fish species (Dept. of Fish & Wildlife), and bottom sediments (Dept. of Ecology). Pesticide analysis has included a number of the WSPMP target compounds (Table 1) except for dacthal, oxadiazon, tetradifon, and the OP pesticides. Pentachlorophenol has been analyzed in place of pentachloroanisole.

Table 5 compares the pesticides and PCBs detected in mussels to results from PSAMP. Relatively few compounds have been identified through analyzing other types of Puget Sound samples. Detections have been limited to fish and sediment, DDT compounds and PCBs almost exclusively. Except for a set of salmon tissues analyzed in 1992, other pesticides (dieldrin and BHC) have been detected in less than 1% of fish samples. PSAMP monitoring stations for fish and sediment are located in offshore areas relatively far from potential sources of pesticides. Sediment detection limits have been higher than in fish and mussel samples.

Although occurring in the same intertidal habitat as mussels, no pesticides or PCBs have been reported from PSAMP's monitoring of littleneck clams. Detection limits have been

Table 5. Pesticides/PCBs Detected (x) in Mussels Compared to Recent PSAMP Findings

Sample Type:	Mussels	Fish*	Sediment	Clams**
Portion:	soft parts	muscle & liver	top 2-cm	soft parts
N = :	6	521	63	117
Date:	1995	1992-94	1992	1992 & 93
Agency:	Ecology	Fish & Wildlife	Ecology	Health
Reference:	(1)	(2)	(3)	(4)
DDT	x	x		
DDE	x	x		
DDD	x	x	x	
dieldrin	x	x		
endosulfan I/II	x			
endosulfan sulfate	x			
cis/trans-chlordane	x			
cis/trans-nonachlor	x			na
heptachlor	x			
heptachlor epoxide	x			
alpha/gamma BHC	x	x		
hexachlorobenzene	x			
dacthal	x	na	na	na
pentachlorophenol/anisole	x			
PCB-1248	x	x		
PCB-1254	x	x	x	
PCB-1260	x	x	x	

* chinook salmon, coho salmon, quillback rockfish, copper rockfish, english sole, & pacific cod
** littleneck clams
na = not analyzed
(1) present study
(2) unpublished WDFW data (Sandra O'Neill)
(3) Dutch et al., 1993
(4) unpublished WDOH data (Glen Patrick)

comparable to those achieved in the mussel samples. Failure to detect these compounds may be partly due to the low lipid content of clams.

Columbia River

The pesticide/PCB data available on the lower Columbia River come from several independent investigations, mentioned below, primarily focused on fish tissue. Several pesticides -- mirex, methylparathion, endrin (aldehyde), malathion, and methoxychlor -- have been detected that were not identified in the Ilwaco mussel sample.

As previously noted, dacthal, heptachlor, and heptachlor epoxide were only detected in mussels from the Columbia River mouth. U.S. Fish & Wildlife Service historical data and recent WSPMP analyses show dacthal levels in upper Columbia River fish (Snake River, Walla Walla River, Columbia R. @ Cascade Locks) rank among the highest statewide or nationally (Schmitt et al., 1985, 1990; Davis, unpublished). Fish tissue data collected in 1991 - 93 indicate dacthal is rarely detectable in the lower river (Tetra Tech, 1995).

Although heptachlor and heptachlor epoxide have been reported in Columbia River fish, numerous analyses have shown that concentrations are not substantially elevated (Schmitt et al., 1985, 1990; EPA, 1992; Tetra Tech, 1995).

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Appendices

Appendix A. Location and Size of 1995 WSPMP Mussel Samples

Site	Date	Latitude (N)	Longitude (W)	Sample No.	N =	Length (mm) (mean+/-2sd)
Padilla Bay (1)	5/17	48 30.4	122 28.9	20-8041	84	31+/-6
Duwamish Waterway (2)	5/17	47 34.4	122 21.2	20-8042	57	40+/-8
Hylebos Waterway (3)	5/17	47 15.6	122 21.4	20-8043	45	45+/-9
Chambers Creek (4)	5/17	47 11.0	122 34.7	20-8044	33	51+/-8
Budd Inlet (5)	5/25	47 02.9	122 53.6	21-8046	30	52+/-7
Columbia R. nr Ilwaco (6)	5/16	46 17.2	124 03.1	20-8040	56	40+/-7

(1) Old pilings near Joe Leary Slough, one mile north of research station
(2) East shore of west waterway, just upstream of Fisher Mills
(3) Railroad bridge pilings at mouth of Hylebos Creek
(4) South shore of creek mouth at Thomas M. Chambers monument
(5) Head of East Bay at culvert at mouth of Moxlie Creek
(6) Fort Canby State Park boat launch

Appendix B

State of Washington Department of Ecology
Manchester Environmental Laboratory
7411 Beach Dr. East Port Orchard WA. 98366
August 23, 1995

Project: WSPMP Fish Tissue

Sample(s): 208040, 208041, 208042, 208043, 208044, 218046

Laboratory: California Department of Fish and Game
Water Pollution Control Laboratory

By: Karin Feddersen

Case Summary

These samples were received at the Manchester Environmental Laboratory on May 26, 1995, and transported to California Department of Fish and Game Water Pollution Control Laboratory on May 31, 1995, for Pest/PCB with subsequent confirmation on GC/MS Ion Trap, % Lipids, and % Non-Polar Lipids analysis.

This data was reviewed for qualitative and quantitative accuracy, validity, and usefulness.

The % Lipids and % Non-Polar Lipids data are acceptable. The Pesticide/PCB data are further reviewed on the following pages.

There is no need to assimilate the "dilution factor" or "sample wt/vol" into the final values reported; these calculations have already been figured into the reported values.

DATA QUALIFIER DEFINITIONS

U - The analyte was not detected at or above the reported result.

J - The associated numerical result is an estimated quantity.

N - The analyte has been tentatively identified.

NJ - The analyte has been tentatively identified. The associated numerical result is an estimated quantity.

C - The presence of the analyte has been positively confirmed on the GC/MS Ion Trap.

JC - The presence of the analyte has been positively confirmed on the GC/MS Ion Trap. The associated numerical result is an estimated quantity.

Holding Times:

A one year holding time from collection to extraction has been established for fish and shellfish in EPA's Guidance For Assessing Chemical Contaminant Data For Use In Fish Advisories. These samples were kept frozen until the time of extraction. The sample extracts were analyzed within forty (40) days from extraction.

Method Blank:

Oxychlordane was detected in the method blank. This analyte was detected in the samples at an amount less than five (5) times that detected in the method blank. The results for oxychlordane in the samples have been qualified with a "U". This qualifier indicates that this analyte was not detected in the samples at or above the suspected laboratory contamination level.

GC/MS Tuning and Calibration:

Calibration against Decafluorotriphenylphosphine (DFTPP) is acceptable for the confirmation of all associated sample extracts.

Calibration:

The % Relative Standard Deviations were within the maximum of 25% for all calibrations.

Duplicates:

A duplicate analysis was performed on sample 208043. Relative Percent Differences (RPD) are reasonable, acceptable, and within advisory QC limits.

Matrix Spikes (MS/MSD):

Sample 218046 was analyzed as a matrix spike and matrix spike duplicate (MS/MSD). Matrix spike recoveries are within method QC limits of $\geq$ 50% of the expected values with several exceptions. Heptachlor recovery was 46% in the MS and 39% in the MSD, indicating a possible low bias for this analyte. Heptachlor is recovered from the first florisil fraction. The surrogate DBOB is also recovered from the first fraction. DBOB recoveries were lower (although still acceptable) in the MS and MSD than in the other samples. Thus the low recovery of heptachlor is most likely specific to the MS and MSD and not indicative of a larger QC problem. Therefore, no qualification is necessary for this analyte.

Endrin Aldehyde, Methyl Parathion, and Ethion recoveries were also low. The corresponding surrogate recoveries for these analytes are comparable with the recoveries in the other samples. These analytes have been qualified; when detected with a "J", when not detected, with a "UJ".

All Relative Percent Differences (RPD) are within method QC limits of 50%.

Surrogates:

All surrogate recoveries for these samples and for the associated method blank are reasonable, acceptable, and within QC limits.

Sample Data:

The RPD between the two columns was greater than 30% for some analytes. When these analytes were not confirmed in the sample by GC/MS, results above the quantitation limit have been qualified with "NJ". Results below the quantitation limit have been changed to "U". This was done in order to be consistent with Manchester Environmental Laboratory's standard reporting format.

All target analytes that have been positively confirmed in the samples by the GC/MS Ion Trap have been qualified with a "C".

This data is acceptable for use as amended.

Location			Ilwaco	Padilla Bay	Duwamish WW		
Sample No.		Method Blank	20-8040	20-8041	20-8042		
Date Extracted	Fresh Weight	6/21/95	6/27/95	6/27/95	6/27/95		
	Quantitation Limit (QL)	Fresh Weight	Fresh Weight	Fresh Weight	Fresh Weight		
	(based on 50g sample weight)	Concentration	Concentration	Concentration	Concentration		
COMPOUND	ug/Kg (ppb)	ug/Kg (ppb)	ug/Kg (ppb)	ug/Kg (ppb)	ug/Kg (ppb)		
aldrin	0.30	U	U	U	U		
cis-chlordane	0.50	U	0.33 J	U	0.97		
trans-chlordane	0.50	U	0.50	U	0.91		
oxychlordane	0.50	0.03 J	U	U	U		
cis-nonachlor	0.75	U	U	U	0.20 J		
trans-nonachlor	0.30	U	U	U	0.66 NJ		
cis-chlordene	0.30	U	U	U	U		
trans-chlordene	0.35	U	U	U	U		
chlorpyrifos	1.5	U	U	U	U		
dicofol	6.5	U	U	U	U		
dichlorobenzophenone	20	U	U	U	U		
dacthal	0.15	U	0.33	U	U		
diazinon	3.0	U	U	U	U		
dieldrin	0.15	U	0.43 J	U	0.22		
endosulfan I	0.18	U	0.18	0.15 J	U		
endosulfan II	0.65	U	U	U	U		
endosulfan sulfate	1.5	U	U	U	U		
endrin	0.10	U	U	U	U		
ethion	3.5	U	UJ	UJ	UJ		
alpha BHC	0.25	U	U	0.06 J	0.06 J		
beta BHC	0.80	U	U	U	U		
gamma BHC	0.40	U	U	U	U		
delta BHC	0.50	U	U	U	U		
2,4'-DDD	1.5	U	U	U	U		
4,4'-DDD	1.5	U	1.8 C	0.05 J	0.88 J		
2,4'-DDE	1.0	U	U	U	U		
4,4'-DDE	0.60	U	6.5 C	0.57 J C	1.4 C		
4,4'-DDMU	1.5	U	U	U	U		
2,4'-DDT	0.65	U	0.25 J	U	0.61 J		
4,4'-DDT	0.80	U	0.91	0.12 J	2.8 NJ		
heptachlor	0.30	U	0.01 J	U	U		
heptachlor epoxide	0.50	U	0.14 J	U	U		
hexachlorobenzene	0.20	U	0.14 J	0.03 J	U		
methoxychlor	3.0	U	U	U	U		
oxadiazon	0.25	U	U	U	U		
ethyl parathion	0.45	U	U	U	U		
methyl parathion	0.30	U	UJ	UJ	UJ		
tetradifon	0.30	U	U	U	U		
toxaphene	15	U	U	U	U		
mirex	0.80	U	U	U	U		
pentachloroanisole	0.55	U	U	U	0.05 J		
endrin aldehyde	0.15	U	UJ	UJ	UJ		
endrin ketone	0.60	U	U	U	U		
PCB 1016	6.0	U	U	U	U		
PCB 1221	6.0	U	U	U	U		
PCB 1232	6.0	U	U	U	U		
PCB 1242	6.0	U	U	U	U		
PCB 1248	6.0	U	U	U	U		
PCB 1254	6.0	U	6.4 N	2 J	32		
PCB 1260	6.0	U	U	U	12 J C		
% Lipid		not analyzed	1.09%	0.846%	1.14%		
% Nonpolar Lipid		not analyzed	0.130%	0.085%	0.102%		
% Moisture		not analyzed	89.0%	89.5%	87.1%		

Location		Hylebos WW	Hylebos WW	Chambers Cr.
Sample No.		20-8043	20-8043 Dup	20-8044
Date Extracted	Fresh Weight	6/29/95	6/29/95	6/27/95
	Quantitation Limit (QL)	Fresh Weight	Fresh Weight	Fresh Weight
	(based on 50g sample weight)	Concentration	Concentration	Concentration
COMPOUND	ug/Kg (ppb)	ug/Kg (ppb)	ug/Kg (ppb)	ug/Kg (ppb)
aldrin	0.30	U	U	U
cis-chlordane	0.50	1.2	1.2	0.37 J
trans-chlordane	0.50	1.0	1.0	U
oxychlordane	0.50	U	U	U
cis-nonachlor	0.75	0.26 J	0.37 J	U
trans-nonachlor	0.30	0.99 NJ	1.1 NJ	0.38
cis-chlordene	0.30	U	U	U
trans-chlordene	0.35	U	U	U
chlorpyrifos	1.5	U	U	U
dicofol	6.5	U	U	U
dichlorobenzophenone	20	U	U	U
dacthal	0.15	U	U	U
diazinon	3.0	U	U	U
dieldrin	0.15	0.71	0.77	0.16 N
endosulfan I	0.15	22	25	U
endosulfan II	0.65	12	13	U
endosulfan sulfate	1.5	7.4	8.4	U
endrin	0.10	U	U	U
ethion	3.5	UJ	UJ	UJ
alpha BHC	0.25	0.10 J	0.12 J	U
beta BHC	0.80	U	U	U
gamma BHC	0.40	0.08 J	0.09 J	U
delta BHC	0.50	U	U	U
2,4'-DDD	1.5	U	U	U
4,4'-DDD	1.5	3.7	3.8	U
2,4'-DDE	1.0	0.14 J	0.15 J	U
4,4'-DDE	0.60	5.9 C	6.4	0.33 J
4,4'-DDMU	1.5	U	U	U
2,4'-DDT	0.65	1.4	1.7	U
4,4'-DDT	0.80	5.5 NJ	5.7 NJ	U
heptachlor	0.30	U	U	U
heptachlor epoxide	0.50	U	U	U
hexachlorobenzene	0.20	0.43	0.45	U
methoxychlor	3.0	U	U	U
oxadiazon	0.25	U	U	U
ethyl parathion	0.45	U	U	U
methyl parathion	0.30	UJ	UJ	UJ
tetradifon	0.30	U	U	U
toxaphene	15	U	U	U
mirex	0.80	U	U	U
pentachloroanisole	0.55	0.28 J	0.31 J	U
endrin aldehyde	0.15	UJ	UJ	UJ
endrin ketone	0.60	U	U	U
PCB 1016	6.0	U	U	U
PCB 1221	6.0	U	U	U
PCB 1232	6.0	U	U	U
PCB 1242	6.0	U	U	U
PCB 1248	6.0	18	17	U
PCB 1254	6.0	42 C	50	6.1
PCB 1260	6.0	5 J	5 J	2 J
% Lipid		1.25%	1.13%	0.958%
% Nonpolar Lipid		0.407%	0.360%	0.128%
% Moisture		85.2%	85.8%	87.2%

Location		Budd Inlet	Budd Inlet	Budd Inlet			
Sample No.		21-8046	21-8046MS	21-8046MSD			
Date Extracted	Fresh Weight	6/21/95	6/21/95	6/21/95			
	Quantitation Limit (QL)	Fresh Weight	Fresh Weight	Fresh Weight			
	(based on 15g sample weight)	Concentration	Concentration	Concentration			
COMPOUND	ug/Kg (ppb)	ug/Kg (ppb)	ug/Kg (ppb)	ug/Kg (ppb)			
aldrin	0.80	U	12	11			
cis-chlordane	1.5	0.49 J	15	14			
trans-chlordane	1.5	0.58	13	12			
oxychlordane	1.5	U	12	11			
cis-nonachlor	2.0	U	22	21			
trans-nonachlor	0.80	0.34	15	16			
cis-chlordene	0.80	U	12	11			
trans-chlordene	0.95	U	14	14			
chlorpyrifos	4.0	U	24	23			
dicofol	20	U	150	130			
dichlorobenzophenone	45	U	35	29			
dacthal	0.40	U	17	16			
diazinon	7.0	U	280	260			
dieldrin	0.35	U	21	19			
endosulfan I	0.50	0.66 NJ	18	17			
endosulfan II	2.0	U	12	16			
endosulfan sulfate	3.0	U	20	24			
endrin	0.30	U	15	14			
ethion	9.0	U	44	54			
alpha BHC	0.70	0.07 J	4.6	4.1			
beta BHC	2.0	U	15	12			
gamma BHC	1.0	U	6.6	5.4			
delta BHC	1.5	U	8.2	5.7			
2,4'-DDD	3.5	U	39	36			
4,4'-DDD	4.0	0.45 J	44	39			
2,4'-DDE	2.0	U	27	28			
4,4'-DDE	2.0	0.70	32	35			
4,4'-DDMU	3.5	U	60	61			
2,4'-DDT	2.0	U	36	37			
4,4'-DDT	2.0	U	99	97			
heptachlor	0.75	U	8.9	7.4			
heptachlor epoxide	1.5	U	12	12			
hexachlorobenzene	0.5	U	7.3	7.2			
methoxychlor	7.5	U	77	76			
oxadiazon	0.65	U	32	30			
ethyl parathion	1.5	U	41	34			
methyl parathion	0.70	UJ	21	18			
tetradifon	0.70	U	36	34			
toxaphene	35	U	U	U			
mirex	2.0	U	50	53			
pentachloroanisole	1.5	0.15 J	11	9.8			
endrin aldehyde	0.35	UJ	7.3	5.9			
endrin ketone	2.0	U	11	13			
PCB 1016	20	U	U	U			
PCB 1221	20	U	U	U			
PCB 1232	20	U	U	U			
PCB 1242	20	U	U	U			
PCB 1248	20	U	U	U			
PCB 1254	20	21	23	24			
PCB 1260	20	U	U	U			
% Lipid		1.44%	1.77%	1.57%			
% Nonpolar Lipid		0.124%	0.126%	0.130%			
% Moisture		84.1%	82.8%	84.0%			

	Percent Recovery	Percent Recovery		Percent Recovery	Percent Recovery			
Sample No.	21-8046MS	21-8046MSD	RPD	21-8046MS*	21-8046MSD*	RPD*		
COMPOUND								
aldrin	58	55	5.0	58	55	5.0		
cis-chlordane	85	79	7.1	85	79	7.1		
trans-chlordane	79	76	3.9	79	76	3.9		
oxychlordane	75	70	8.0	75	70	8.0		
cis-nonachlor	86	83	4.0	86	83	4.0		
trans-nonachlor	71	78	9.4	71	78	9.4		
cis-chlordene	59	56	4.7	59	56	4.7		
trans-chlordene	61	59	2.8	61	59	2.8		
chlorpyrifos	52	51	1.6	52	51	1.6		
dicofol	106	91	14.0	106	91	14.0		
dichlorobenzophenone	92	78	16.0	92	78	16.0		
dacthal	77	70	9.7	72	68	5.7		
diazinon	62	58	6.0	62	58	6.0		
dieldrin	92	87	5.0	92	87	5.0		
endosulfan I	89	86	3.6	86	83	3.6		
endosulfan II	80	78	2.2	80	78	2.2		
endosulfan sulfate	73	64	13.0	73	64	13.0		
endrin	89	85	4.9	89	85	4.9		
ethion	39	49	21.5	39	49	21.5		
alpha BHC	55	50	8.7	55	50	8.7		
beta BHC	57	46	21.8	57	46	21.8		
gamma BHC	54	45	18.3	54	45	18.3		
delta BHC	51	35	36.2	51	35	36.2		
2,4'-DDD	88	83	5.7	88	83	5.7		
4,4'-DDD	96	86	11.5	96	86	11.5		
2,4'-DDE	68	73	7.0	68	73	7.0		
4,4'-DDE	80	87	8.4	80	87	8.4		
4,4'-DDMU	74	77	4.3	74	77	4.3		
2,4'-DDT	83	86	3.6	83	86	3.6		
4,4'-DDT(F1 Mix & F2 Mix)	94	93	1.4	94	93	1.4		
heptachlor	46	39	17.4	46	39	17.4		
heptachlor epoxide	75	70	7.5	75	70	7.5		
hexachlorobenzene	57	56	2.1	57	56	2.1		
methoxychlor	85	83	2.3	85	83	2.3		
oxadiazon	81	76	5.4	81	76	5.4		
ethyl parathion	57	47	18.4	55	45	20.0		
methyl parathion	47	41	13.9	47	41	13.9		
tetradifon	81	71	7.4	81	71	7.4		
mirex	96	102	6.0	96	102	6.0		
pentachloroanisole	60	54	9.4	60	54	9.4		
endrin aldehyde	39	30	25.0	37	29	24.2		
endrin ketone	74	66	11.7	74	66	11.7		
DBOB (F1,F2, & F3 Mix)	51	50	3.3	51	50	3.3		
DCB (F1,F2, & F3 Mix)	98	98	0.1	98	98	0.1		
DBCE (F1,F2, & F3 Mix)	87	83	5.3	87	83	5.3		
average	72	69	9.0	72	69	9.0		
std. dev.	17	18	7.3	17	18	7.3		
* Recoveries corrected for analyte or contamination reported in unspiked sample								

Sample No.	Percent DBOB Recovered		Percent DCB Recovered		Percent DBCE Recovered
Method Blank	64		100		110
21-8046	63		110		82
21-8046MS	51		98		87
21-8046MSD	50		98		83
21-8040	73		97		92
21-8041	75		100		73
21-8042	72		110		94
21-8043	75		93		89
21-8043 dup.	74		96		87
20-8044	72		100		90
average	67		100		89
std. dev.	9.6		5.7		9.4

DBOB – 4,4-dibromooctafluorobiphenyl

DCB – decachlorobiphenyl

DBCE – dibutylchlorendate

Appendix C. Historical Data on Pesticides/PCBs in Washington Mussels (ug/Kg (ppb) wet wt.)

Compound	Year	Elliott Bay	Commencement Bay	Budd Inlet	Columbia River
t-DDT	1975[1]	na	na	na	na
	1976[2]	na	na	na	0.3*
	1977[2]	na	na	na	0.8*
	1978[2]	na	na	na	na
	1981[3]	na	6.8	na	na
	1982[3]	na	nd	na	na
	1986[4]	17	4.3	1.8	9.8
	1987[4]	4.6	2.6	2.6	4.7
	1988[4]	6.4	0.8	3.2	8.2
	1995[5]	5.1	16	1.2	3.4
dieldrin	1981	na	nd	na	na
	1982	na	nd	na	an
	1986	1.6	0.7	0.6	0.6
	1987	0.3	1.8	0.9	0.9
	1988	0.4	nd	0.4	0.5
	1995	nd	0.1	nd	0.4
t-chlordane**	1981	na	nd	na	na
	1982	na	nd	na	na
	1986	3.5	1.8	0.9	1
	1987	0.8	2.5	1.7	0.6
	1988	1.3	0.2	1.4	0.8
	1995	1.2	1.5	0.5	0.5
gamma BHC	1981	na	nd	na	na
	1982	na	nd	na	na
	1986	0.04	0.2	0.2	0.1
	1987	nd	0.09	0.2	0.04
	1988	0.2	0.02	0.1	0.1
	1995	nd	0.08	nd	nd
hexachlorobenzene	1981	na	nd	na	na
	1982	na	nd	na	na
	1986	nd	nd	nd	nd
	1987	nd	0.09	0.04	nd
	1988	0.3	0.4	0.2	0.1
	1995	nd	0.4	nd	0.1

Appendix C. (continued)

Compound	Year	Elliott Bay	Commencement Bay	Budd Inlet	Columbia River
t-PCBs	1975[1]	210	72	27	na
	1976[2]	na	na	na	5
	1977[2]	na	na	na	3
	1978[2]	na	na	na	3
	1981[3]	na	82	na	na
	1982[3]	na	26	na	na
	1986[4]	143	25	17	11
	1987[4]	75	44	22	12
	1988[4]	58	5	18	14
	1995[5]	44	70	21	6

na = not analyzed
nd = not detected
* DDE only
** t-chlordane = alpha-chlordane + trans-nonachlor + heptachlor + heptachlor epoxide (as reported in NOAA, 1989)
(1) Mowrer et al., 1977 [Elliott Bay site is W. Duwamish WW; Commencement Bay site is mouth of Hylebos WW; Budd Inlet site is Priest Point Park]
(2) Farrington et al., 1982 [Columbia River site is North Jetty: *Mytilus californianus*]
(3) Hopkins et al., 1985 [Commencement Bay site is mouth of City Waterway]
(4) NOAA ,1989 [Elliott Bay site is Four-Mile Rock; Commencement Bay site is Tahlequah Pt.]
(5) present study [Elliott Bay site is W. Duwamish WW; Commencement Bay site is head of Hylebos WW]
Note: Data from references (2) and (4) converted from dry wt. to wet wt. assuming 87% moisture (Table 2)