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Results of a Screening Analysis for Metals and Organic Compounds in Shellfish from Padilla Bay and Vicinity

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Results of a Screening Analysis for Metals and Organic Compounds in Shellfish from Padilla Bay and Vicinity

by
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Prepared for and in cooperation with the Swinomish Tribal Community, Skagit County Health Department, Washington State Department of Fish and Wildlife Spills Response Team, and the Washington State Department of Ecology Spills Prevention, Preparedness, and Response Program.

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Abstract

Fourteen composite edible tissue samples from crabs, clams, oysters, and mussels collected in the Padilla Bay area were screened for over 130 potentially toxic and bioaccumulative metals and organic compounds, in response to health concerns expressed by the Swinomish Tribal Community. An extensive list of polyaromatic hydrocarbons (PAH) were included in the analyses to provide baseline data for state agency spills programs in the event of oil spills in the area.

Results showed little or no evidence of significant contamination. Slight elevations in several chemicals, including lead, tributyltin, DDT compounds, and PAH, were observed in areas known or suspected to have sources of these compounds. The Washington State Department of Health reviewed the data and concluded that most chemicals were at levels below human health concerns. Although arsenic and 2,3,7,8 -tetrachlorodibenzofuran concentrations appeared to be at background levels for Puget Sound, the concentrations in the shellfish samples exceeded human health screening levels. The Department of Health recommended additional sampling for arsenic.

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Summary

The Washington State Department of Ecology (Ecology) analyzed shellfish from Padilla Bay, Fidalgo Bay, the Swinomish Channel, and Samish Island (a reference area) for over 130 potentially toxic and bioaccumulative chemicals that could pose a human health concern. The species tested were Dungeness crabs, littleneck clams, butter clams, Japanese oysters, and bay mussels. This was a screening survey to determine if there was a need for more intensive follow-up sampling.

Fourteen composite edible tissue samples were analyzed, as indicated below (number of individual organisms in each composite shown):

Location	Dungeness Crab	Littleneck Clams	Japanese Oyster	Butter Clams	Mussels
Fidalgo Bay/Crandall Spit	5	50			
W. Padilla Bay/NE March Point	5*	50			50**
Swinomish Channel			20		
E. Padilla Bay/Person Road		50			
E. Padilla Bay/Bayview St. Park			20		
Hat Island	5				
Samish Island (reference area)	5*			50	50**

*   hepatopancreas also analyzed, but for PAH only
** analyzed for PAH only

The study was conducted at the request of the Swinomish Tribal Community. The Tribe was concerned about the potential for chemical contamination of food species due to discharges from March Point refineries, agriculture, marinas, boatyards, and other sources. At the request of the Ecology Spills Prevention, Preparedness, and Response Program and the Washington State Department of Fish and Wildlife Habitat Program, Spills Response Team, the analysis included a range of polyaromatic hydrocarbons (PAH). The PAH data were needed to establish a baseline in the event of oil spills. Data from this project were reviewed by the Washington State Department of Health (WDOH), who provided consultation regarding the need for further actions to ensure protection of human health.

The following chemicals were analyzed:

- Arsenic
- Lead
- Cadmium
- Selenium
- Mercury
- Tributyltins
- Polyaromatic Hydrocarbons
- Polychlorinated Biphenyls
- Bioaccumulative Pesticides
- Polychlorinated Dioxins and –Furans

The analysis of crab muscle showed the highest metals concentrations were for arsenic at 5,230 - 8,390 ug/Kg (parts per billion, wet weight), followed by selenium at 496 - 692 ug/Kg, mercury at 41 - 75 ug/Kg, and lead at 11 - 33 ug/Kg. Cadmium was not detected at or above 25 ug/Kg in any of the crab samples.

A different pattern was seen in clams and oysters which had 1,360 - 2,600 ug/Kg of arsenic, 211 - 1,440 ug/Kg of cadmium, 470 - 750 ug/Kg of selenium, 43 - 128 ug/Kg of lead, and 11 - 26 ug/Kg of mercury.

Except for lead in littleneck clams collected at Crandall Spit and at the end of March Point, metals concentrations were not substantially different from those in crabs and clams from the Samish Island reference area.

Only a few organic compounds were detected in crab muscle. Two or three samples had low levels of the DDT breakdown product DDE at 0.19 - 0.25 ug/Kg, PCB-1248 at 1.2 - 1.4 ug/Kg, and 2,3,7,8-tetrachlorodibenzofuran (TCDF) at 0.49 - 0.51 ng/Kg (parts per trillion). PAH and tributyltins were not detectable in crab muscle, except for a trace, 4.9 ug/Kg, of naphthalenes in the reference area crabs.

Many more organic compounds were detected in clams and oysters than in crabs, particularly in oysters. No organic compounds were detectable in the reference area butter clam sample.

Tributyltin, pesticides, TCDF, and, with one exception, PCBs were found in oysters but not clams. Tributyltin was higher in the Swinomish Channel oysters than the Bayview State Park oysters, 7.9 and 2.7 ug/Kg, respectively. The Bayview oysters had slightly higher concentrations of DDT compounds, 2.2 vs. 1.1 ug/Kg, and this was the only location where dieldrin was detectable, at 0.25 ug/Kg. PCB and TCDF concentrations were similar in the two oyster samples and comparable to the levels observed in crab muscle. The lone detection of PCBs in clams was 2.3 ug/Kg of PCB-1248 in the March Point littlenecks.

PAH concentrations in the clams were low to non-detectable at March Point, Person Road, and Samish Island. Higher concentrations occurred in the Crandall Spit clam sample. The largest number of PAH compounds were quantified in the Swinomish Channel oyster and the March Point mussels. Concentrations of individual PAH at these two sites were in the range of 2.5 - 32 ug/Kg. Almost no PAH were detectable in shellfish from the Samish Island reference area.

Results from this study show there is a low level of contamination in the study area relative to other parts of Puget Sound. Although the findings are unremarkable, they are consistent with known or suspected sources of contamination. For example: (1) Elevated tributyltin and PAH in the Swinomish Channel potentially due to the concentration of marinas, boatyards, and vessel traffic, (2) Elevated lead and PAH in the March Point area potentially due to road runoff, stormwater, wastewater treatment plant effluent, refinery effluents/flare towers, and vessel traffic, and (3) Pesticides detected on the east shore of Padilla Bay potentially due to historical use on nearby agricultural land.

Where sufficient numbers of PAH were quantified to see a distribution pattern, combustion sources were indicated for the March Point area and petroleum as the source in the Swinomish Channel. Only methylnaphthalenes were detected in crab tissues, primarily in hepatopancreas, suggesting petroleum, probably from a variety of origins, as the source.

WDOH compared the detected chemical concentrations with human health screening values for each of the species tested, using shellfish consumption rates determined for the Tulalip Tribe. Only two chemicals, arsenic and TCDF, exceeded screening levels, indicating the need for a more detailed evaluation in order to determine the human health implications of these chemicals at the concentrations detected. Although the arsenic concentrations were generally comparable to shellfish from other parts of Puget Sound, lack of data on the chemical form of arsenic, and therefore toxicity, in shellfish leads to uncertainty in conclusions about the presence of a health risk. WDOH recommended that further sampling and analysis be conducted for total and inorganic arsenic in frequently consumed shellfish species from the study area. No additional sampling was recommended for TCDF or other dioxins and furans because of the background level of the concentrations found and limitations of current toxicological understanding of these compounds.

Acknowledgements

The people listed below advised on the design of this study and/or assisted with collection of samples. Their generous help is very much appreciated.

◊ Swinomish Tribal Community: Aundrea Noffke, John Stone, Joe McDonald, Abe McDonald

◊ Skagit County Health Department: Ken Willis, John Hadman, Andy Ross

◊ Washington State Department of Health: Glen Patrick, Joan Hardy

◊ Washington State Department of Fish and Wildlife: Dan Doty, Charmane Ashbrook, Mark O'Toole

◊ Washington State Department of Ecology: Doug Bulthuis, Dale Davis, Jerry Shervey, Dave Rogowski, Dave Serdar

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The final report was formatted and proofread by Shirley Rollins and Joan LeTourneau.

Introduction

In response to a request from the Swinomish Tribal Community, the Washington State Department of Ecology (Ecology) Environmental Assessment Program (EAP) analyzed shellfish from Padilla Bay and adjacent waters for chemical contaminants that could pose a human health risk. Contaminant sources of concern to the Tribe included petroleum refineries and other industry on March Point, Skagit Valley agriculture, the abandoned Whitmarsh Landfill in Padilla Bay Lagoon, marinas and boatyards at Anacortes and La Conner, and vessel traffic.

The Ecology Spills Prevention, Preparedness, and Response Program and the Washington State Department of Fish and Wildlife Habitat Program, Spills Response Team, funded an expanded analysis for polyaromatic hydrocarbons (PAH) in the shellfish samples. Information on the concentrations and relative abundance of PAH in Padilla Bay and in background areas was needed for resource damage assessments in the event of oil spills.

The data obtained through this study were provided to the Washington State Department of Health (WDOH) for review and consultation regarding the need for further actions to ensure protection of human health. Their conclusions and recommendations are contained at the end of this report.

Only limited chemical analyses have been conducted on shellfish from the Padilla Bay area. Results, discussed later in this report, have suggested a generally low level of contamination. However, data from sediment and water monitoring programs point to several locations as sources of bioaccumulative chemicals.

Anacortes

Sediments in Anacortes marinas and navigation channels have elevated concentrations of cadmium and lead, as well as the antifouling agent tributyltin. Some sediment samples have exceeded state standards for PAH (Crecelius, 1986; Crecelius et al., 1989; Tetra Tech, 1991). The nearshore area adjacent to the old Scott Paper mill is heavily impacted with wood, brick, and other debris from past operations. There are traces of dioxin and PCBs in the sediments (Turvey, 1999).

March Point

Persistent bioaccumulative chemicals that could potentially be discharged from the March Point refineries include cadmium, mercury, PAH, and polychlorinated dioxins and -furans. Arsenic and lead are present in the effluents but usually below limits of detection (Wigfield, 1999). Elevated levels of high molecular weight PAH occur in the sediments around March Point. Sediments near the refinery outfalls have exceeded standards for cadmium and the PAHs phenanthrene and fluoranthene (Barrick and Prahl, 1987; Johnson et al., 1997).

The March Point wastewater treatment plant discharges to inner Fidalgo Bay just south of Crandall Spit. Stormwater also discharges there.

Up to 20,000 gallons of North Slope crude were spilled to inner Fidalgo Bay in February 1991. In surveys conducted for the Department of Natural Resources, EAP collected sediment samples in 1997 and 1999 to determine if widespread contamination still existed (Johnson et al., 1997; Johnson, 2000). With the exception of one intertidal area inside Crandall Spit, contaminated with oil and with PAH exceeding standards, there was little evidence of significant contamination.

Padilla Bay Lagoon

Petroleum, PCBs, PAH, and other contaminants have been detected in seepage to this lagoon from the abandoned Whitmarsh Landfill at the head of Padilla Bay. Lagoon sediments exceed cleanup screening levels for methylphenols and sediment bioassays, with several sites also showing some elevations in polychlorinated dioxins and -furans. Extremely high levels of petroleum are found in one part of the lagoon (Johnson, 1999).

Padilla Bay Sloughs

A number of pesticides, herbicides, and breakdown products have been detected in water samples from sloughs draining Skagit County agricultural lands on the eastern shore of Padilla Bay (Mayer and Elkins, 1990; Davis and Johnson, 1994; Davis, 1996). Detections have included diazinon, dacthal, pentachlorophenol, mevinphos, 3-hydroxycarbofuran, 2,4-D, dicamba, chlorpropham, bentazon, eptam, MCPA, MCPP, and triclopyr. Among these compounds, diazinon, dacthal, and pentachlorophenol can accumulate in shellfish.

Study Plan

Objectives

A Tier 1 screening study was conducted as described in EPA (1995) *Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories*. The objective of this type of study is to identify areas where fish or shellfish are frequently harvested and where concentrations of chemical contaminants exceed specified human health screening values, thus requiring more intensive follow-up sampling or chemical-specific toxicity evaluations. A secondary study objective was to obtain PAH data for the Ecology and Fish & Wildlife spills programs.

Species and Locations

The species used for the human health assessment were Dungeness crab (*Cancer magister*), native littleneck clams (*Prototheca staminea*), Japanese oysters (*Crassostrea gigas*), and butter clams (*Saxidomus giganteus*). These are the shellfish most heavily consumed from the study area. Muscle tissue was analyzed from crabs, and the entire soft parts from clams and oysters.

Six harvest areas were selected with the advice of the Swinomish Tribal Community, Skagit County Health Department, Skagit System Cooperative, and Padilla Bay National Estuarine Research Reserve (Figure 1). These included both tribal and recreational sites.

Crab samples were obtained from three areas: in Fidalgo Bay off Crandall Spit, in Padilla Bay off the end of March Point, and near Hat Island. Littlenecks are the most abundant clams in the study area and were collected in Padilla Bay at Person Road north of Bayview, at the northeast tip of March Point, and on Crandall Spit. Oysters were collected at Bayview State Park and on reservation land along the west shore of the Swinomish Channel.

Shellfish from a reference location, Samish Island, about five miles north of March Point, were sampled to give a baseline for the chemicals of interest. Samish Island is an established sediment reference area known to have a low level of chemical contaminants (PTI, 1991b). Littleneck clams collected to the east in Samish Bay for the Puget Sound Ambient Monitoring Program (PSAMP) have shown no significant chemical contamination (Patrick, 1996). There is subsistence harvest of clams along the south shore of Samish Island (Noffke, 1999). Reference samples included crabs and butter clams. The number of littleneck clams encountered here was too low for sampling.

Several additional samples were analyzed for PAH at the request of agency spills programs. These included bay mussels (*Mytilus* sp.) from the refinery wharves on March Point and at Samish Island, as well as hepatopancreas tissue from the crabs collected off March Point and Samish Island.

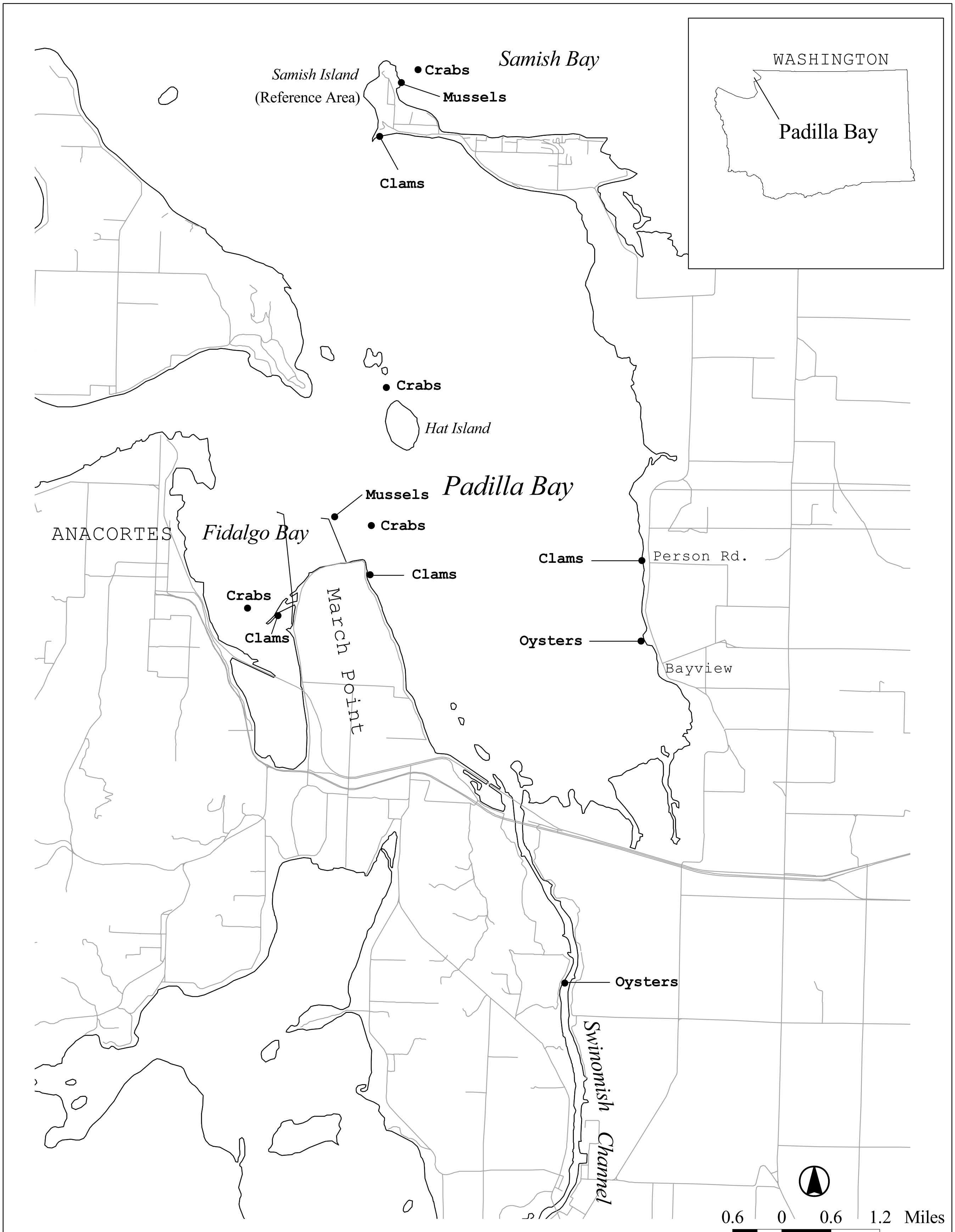

Figure 1. Shellfish Sampling Sites, May - June 1999

Sample Size

As recommended by EPA (1995), composite samples were analyzed to provide a conservative estimate of typical exposures. Composite samples are a cost-effective way to estimate average tissue concentrations. For each sampling site, a single composite sample was prepared for each species.

The crab composites consisted of pooled muscle or hepatopancreas tissue from five legal-size males (6 ¼ inch carapace width). A sample size of five has been used in other studies on chemical contaminants in Puget Sound crabs (PTI, 1991a) and is commonly used in fish tissue studies (EPA, 1992).

Each clam and mussel sample consisted of the pooled entire soft parts from approximately 50 individual organisms. PSAMP has used a sample size of 30 for monitoring chemicals in littleneck and butter clams (Patrick, 1996). Twenty individuals were used for the oyster composites. All the clams taken for analysis were legal size (> 1 ½ inch; there is no size limit for oysters or mussels) and all specimens were unbroken.

Table 1 shows the species and number of individuals analyzed from each sampling site. Appendix A has detailed information on sampling locations and the size of the organisms included in the composites.

Table 1. Locations, Species, and Number of Individual Organisms Analyzed.

Location	Dungeness Crab	Littleneck Clams	Japanese Oyster	Butter Clams	Mussels
Fidalgo Bay/Crandall Spit	5	50			
W. Padilla Bay/NE March Point	5	50			50
Swinomish Channel			20		
E. Padilla Bay/Person Road		50			
E. Padilla Bay/Bayview St. Park			20		
Hat Island	5				
Samish Island	5			50	50

Timing

The crab samples were collected May 26, 1999 with the assistance of the Swinomish Tribe. Most of the subsistence harvest of crabs by the Swinomish begins in early May and continues until October, but can occur throughout the year (Noffke, 1999). The recreational season for crabs is year-round, except pots are restricted to July 16 - April 15. The commercial crabbing season begins in late May.

Clam, oyster, and mussel samples were collected during the low tide of June 1, 1999. The Skagit County Health Department, Swinomish Tribe, and representatives of the agency spills programs helped with the collection. Most of the subsistence harvest of clams begins in May and continues into the fall as long as daylight low tides allow, but can occur throughout the year (Noffke, 1999). Recreational harvest of clams, oysters, and mussels is open year-round. PSAMP clam samples have been collected in March or May when lipid reserves are highest, prior to the summer spawning season (Patrick, 1996). Because many of the chemicals being analyzed are lipid (fat) soluble, a spring/early summer sampling period increases the likelihood of their detection.

Target Chemicals

Chemicals analyzed in the shellfish samples are listed below. The number of compounds or chemical mixtures determined are shown in parentheses. Table 2 contains a detailed listing.

Arsenic	Polyaromatic Hydrocarbons (55+)
Lead	Polychlorinated Biphenyls (7)
Cadmium	Bioaccumulative Pesticides (44)
Selenium	Polychlorinated Dioxins and –Furans (17)
Mercury	Percent Lipids
Tributyltins (4)	Percent Solids

The analyses included all but three of the chemicals recommended by EPA (1995) for fish consumption studies. Due to lack of an adequate analytical method for tissue, the EPA pesticides disulfoton, terbufos, and oxyflourfen were not analyzed. There are no reports of the occurrence of these compounds in Puget Sound biological samples. They are rarely or never detected in state surface waters (Davis, 1998).

Data were obtained on chemicals beyond those recommended by EPA. These included 18 additional bioaccumulative pesticides. A number of these have either been detected in Puget Sound shellfish – endosulfan sulfate, alpha BHC, dacthal, pentachloroanisole (metabolite of pentachlorophenol) – and/or water samples from Puget Sound tributaries – parathion, chlorpyrifos, pentachlorophenol, dacthal (Davis, 1998; Johnson and Davis, 1996). Over 40 additional PAH compounds were analyzed. The extensive PAH analysis (National Oceanic Atmospheric Administration [NOAA] list) was intended to establish a baseline for PAH in the study area and permit differentiation between PAH from petroleum and other sources (e.g., combustion of fossil fuels or creosote).

Percent lipid was analyzed for use in normalizing the organics data. Percent solids was also determined.

Table 2. Chemicals Analyzed in Shellfish Samples.

CAS No.	Chemical Name	CAS No.	Chemical Name
Metals		**Polyaromatic Hydrocarbons**	
7440382	Arsenic	3001950	C1-Naphthalenes
7439921	Lead	3001951	C2-Naphthalenes
7440439	Cadmium	3001952	C3-Naphthalenes
7782492	Selenium	3001953	C4-Naphthalenes
7439976	Mercury	3001960	C1-Phenanthrenes/Anthracenes
		3001961	C2-Phenanthrenes/Anthracenes
Polyaromatic Hydrocarbons		3001962	C3-Phenanthrenes/Anthracenes
50328	Benzo[a]pyrene*	3001963	C4-Phenanthrenes/Anthracenes
53703	Dibenzo[a,h]anthracene*	3001954	C1-Fluorenes
56553	Benzo[a]anthracene*	3001955	C2-Fluorenes
83329	Acenaphthene	3001956	C3-Fluorenes
85018	Phenanthrene	3001957	C1-Dibenzothiophenes
86737	Fluorene	3001958	C2-Dibenzothiophenes
90120	1-Methylnaphthalene	3001959	C3-Dibenzothiophenes
91203	Naphthalene	3001964	C1-Fluoranthene/Pyrene
91576	2-Methylnaphthalene	3001965	C1-Chrysenes
91587	2-Chloronaphthalene	3001966	C2-Chrysenes
92524	1,1'-Biphenyl	3001967	C3-Chrysenes
120127	Anthracene	3001968	C4-Chrysenes
129000	Pyrene		
132649	Dibenzofuran	**Polychlorinated Dibenzo-p-dioxins**	
132650	Dibenzothiophene	1746016	2,3,7,8-TCDD*
191242	Benzo[ghi]perylene	40321764	1,2,3,7,8-PeCDD*
192972	Benzo[e]pyrene	39227286	1,2,3,4,7,8-HxCDD*
193395	Indeno[1,2,3-cd]pyrene*	57652857	1,2,3,6,7,8-HxCDD*
198550	Perylene	19408743	1,2,3,7,8,9-HxCDD*
205992	Benzo[b]fluoranthene*	35822469	1,2,3,4,6,7,8-HpCDD*
206440	Fluoranthene	3268879	OCDD*
207089	Benzo[k]fluoranthene*		
208968	Acenaphthylene	**Polychlorinated Dibenzofurans**	
218019	Chrysene*	51207319	2,3,7,8-TCDF*
483658	Retene	57117416	1,2,3,7,8-PeCDF*
581420	2,6-Dimethylnaphthalene	57117314	2,3,4,7,8-PeCDF*
832699	1-Methylphenanthrene	70648269	1,2,3,4,7,8-HxCDF*
1576676	Phenanthrene, 3,6-dimethyl	57117449	1,2,3,6,7,8-HxCDF*
1730376	9H-Fluorene, 1-methyl	60851345	2,3,4,6,7,8-HxCDF*
2245387	1,6,7-Trimethylnaphthalene	72918219	1,2,3,7,8,9-HxCDF*
2531842	2-Methylphenanthrene	67562394	1,2,3,4,6,7,8-HpCDF*
3698243	Chrysene, 5-methyl	55673897	1,2,3,4,7,8,9-HpCDF*
- -	4,6-Dimethyldibenzothiophene	39001020	OCDF*

Table 2. Chemicals Analyzed in Shellfish Samples (cont'd).

CAS No.	Chemical Name	CAS No.	Chemical Name
Butyltins		**Bioaccumulative Pesticides**	
1461252	Tetrabutyltin	959988	Endosulfan I*
56573854	Tributyltin*	1022226	DDMU
1002535	Dibutyltin	1024573	Heptachlor Epoxide*
78763549	Monobutyltin	1031078	Endosulfan Sulfate
		1582098	Treflan (Trifluralin)
Polychlorinated Biphenyls		1825214	Pentachloroanisole
11096825	PCB - 1260*	1861321	DCPA (dacthal)
11097691	PCB - 1254*	1966309	Oxadiazon
11104282	PCB - 1221*	2385855	Mirex*
11141165	PCB - 1232*	2921882	Chlorpyriphos*
12672296	PCB - 1248*	3424826	2,4'-DDE
12674112	PCB - 1016*	5103719	Cis-Chlordane (Alpha-Chlordane)*
53469219	PCB - 1242*		
		5103731	Cis-Nonachlor*
Bioaccumulative Pesticides		5103742	Trans-Chlordane (Gamma)*
50293	4,4'-DDT	7421934	Endrin Aldehyde
53190	2,4'-DDD	8001352	Toxaphene*
56382	Ethylparathion	27304138	Oxychlordane*
58899	Gamma-BHC (Lindane)*	33213659	Endosulfan II*
60571	Dieldrin*	39765805	Trans-Nonachlor*
72208	Endrin*	53494705	Endrin Ketone
72435	Methoxychlor	56534022	Alpha-Chlordene*
72548	4,4'-DDD	56641384	Gamma-Chlordene*
72559	4,4'-DDE		
76448	Heptachlor		
90982	Dichlorobenzophenone		
115322	Kelthane (Dicofol)*		
116290	Tetradifon		
118741	Hexachlorobenzene*		
298000	Methyl Parathion		
309002	Aldrin		
319846	Alpha-BHC		
319857	Beta-BHC		
319868	Delta-BHC		
333415	Diazinon*		
563122	Ethion*		
789026	2,4'-DDT		

* recommended in EPA (1995)

Field Sampling and Tissue Preparation

Crabs

Crab sampling and tissue preparation procedures were based on PTI (1991a) and EPA (1996).

Dungeness crabs were collected using pots baited with squid and mackerel, and set overnight. Only male crabs with carapace widths greater than the 6 ¼ inch legal limit were taken. Care was taken to avoid having the crabs come in contact with engine fumes, fuel, oil, bilge water, or other contaminants. Sampling site coordinates were recorded from a hand-held Global Positioning System (GPS). An Ecology representative was on hand when the pots were hauled, and he packaged the samples immediately.

Each crab selected for analysis was killed with a blow to the ventral nerve cord. The crabs were individually wrapped in aluminum foil, put in double plastic bags, labeled with date and location of collection, and placed in coolers containing blue ice. The crabs were kept shell side down so body cavity fluids drained away from muscle tissue. The samples were transported to the Ecology Headquarters chain-of–custody room within one day of collection and were frozen in a secure freezer.

Muscle and hepatopancreas tissues were resected from the crabs using techniques to minimize potential for sample contamination. Only non-corrosive stainless steel instruments were used. People preparing the samples wore non-talc polyethylene gloves and worked on aluminum foil. The gloves and foil were changed between samples.

The carapace width of each crab was recorded to the nearest 1/8 inch. After rinsing the shell with tap water and de-ionized water to remove any adhering debris, muscle tissue was removed from the legs, claws, and body and placed in glass jars with teflon lid-liners, cleaned to EPA (1990) QA/QC specifications. For the March Point and reference area crabs, hepatopancreas tissue was removed similarly. Care was taken not to include hepatopancreas tissue in the muscle samples. Shell fragments were not included in the samples.

Tissues from five individual crabs were composited for each sampling site, using equal tissue weights from each individual. The resected samples were homogenized to uniform color and consistency in a plastic and stainless steel Kitchen-Aid blender. The remaining crabs were kept frozen at Ecology Headquarters in the event that more tissue was needed for analysis.

Cleaning of resecting instruments and blender parts was done by washing in tap water with Liquinox detergent, followed by sequential rinses with tap water, de-ionized water, and pesticide-grade acetone. The items were then air dried on aluminum foil in a fume hood before use.

Subsamples of the muscle and hepatopancreas homogenates were split into new glass jars with teflon lid liners, cleaned to EPA specifications. The samples were refrozen and taken by courier to the Ecology Manchester Environmental Laboratory (Manchester) on June 3, 1999. They were stored frozen at Manchester until analyzed. Containers with excess samples were stored frozen at Ecology Headquarters.

Chain-of-custody was maintained throughout the above procedure.

Clams, Oysters, and Mussels

Clam, oyster, and mussel sampling and tissue preparation procedures were based on unpublished guidelines prepared by Glen Patrick, Washington State Department of Health, Office of Environmental Health Assessments. These are modifications of procedures used for PSAMP shellfish monitoring.

Approximately 80 individual clams and mussels, and 30 oysters, were collected at each site. The clams and oysters were taken from within a 100 ft. stretch of beach. The March Point mussels were collected from concrete pilings at the end of the Tesoro refinery wharf. The Samish Island mussel sample was collected from rocks. Sampling site coordinates were recorded from a hand-held GPS. An Ecology representative was present at all sampling sites.

Clam diggers used clean rakes or shovels, uncontaminated with grease or oil. The shellfish were placed in plastic buckets. Where gloves were used for sample collection, these were newly purchased for the study. Rakes, shovels, buckets, and gloves were washed with seawater between sampling sites.

The shellfish were rinsed thoroughly with seawater to remove adhering mud and sand, then placed in one-gallon glass jars with teflon lid-liners, cleaned to EPA specifications. Each jar was labeled with date and location of collection, wrapped in bubble-wrap to avoid breakage, and placed in coolers containing blue ice. The samples were transported to the Ecology Headquarters chain-of–custody room within one day of collection and frozen in a secure freezer.

Tissues were removed using techniques to minimize potential for sample contamination. Only non-corrosive stainless steel instruments were used. Non-talc polyethylene gloves were worn and the work was done on aluminum foil. The gloves and foil were changed between samples.

The range (minimum and maximum) of shell widths (clams) or lengths (oysters/mussels) for the organisms being included in each composite were recorded to the nearest 1/16 inch. The composites included approximately equal numbers of small, medium, and large individuals. Shell fragments and mussel byssal threads were not included in the samples.

After rinsing the shellfish with tap water and de-ionized water to remove any remaining debris, the entire soft parts were removed and placed in glass jars, cleaned to EPA specifications. The tissues from 50 individual clams and mussels, and 20 oysters, were composited for each sampling site. The soft parts were homogenized to uniform color and consistency in a plastic and stainless steel Kitchen-Aid blender. The remaining shellfish were kept frozen at Ecology Headquarters.

Cleaning of resecting instruments and blender parts was done by washing in tap water with Liquinox detergent, followed by sequential rinses with tap water, de-ionized water, and pesticide-grade acetone. The items were air dried on aluminum foil in a fume hood before use.

Subsamples of the homogenates were split out into new glass jars, cleaned to EPA specifications. The samples were refrozen and taken by courier to Manchester Laboratory on June 3, 1999. They were stored frozen at Manchester until analyzed. Containers with excess sample were stored frozen at Ecology Headquarters.

Chain-of-custody was maintained throughout the above procedure.

Analytical Methods

Methods used for analyzing the shellfish samples are shown in Table 3. The work was done by Manchester Laboratory, except for polychlorinated dioxins and -furans which were analyzed by Pace Analytical in Minneapolis, Minnesota, a contractor selected by Manchester. The PAH and dioxin/furan data are recovery corrected.

Table 3. Analytical Methods for Shellfish Samples.

Analysis	Method
Mercury	CVAA, EPA Method 245.5
Lead	ICP/MS, EPA Method 200.8
Arsenic	ICP/MS, EPA Method 200.8
Cadmium	ICP/MS, EPA Method 200.8
Selenium	ICP/MS, EPA Method 200.8
Tributyltins	GC/AED, EPA Method 3545
Pesticides/PCBs	GC/ECD, EPA Methods 3540, 3620, 3665, 8082*
Polychlorinated Dioxins/Furans	High Res. GC/MS, EPA Method 8290**
Polyaromatic Hydrocarbons	GC/SIM-MS, EPA Method 8270*
Percent Lipid	Gravimetric, EPA Method 608.5
Percent Solids	Gravimetric, EPA Method 160.3

* as modified by Manchester
** with enhancements from EPA Method 1613B

Data Quality

QA Review

Manchester staff prepared written reviews on the quality of the chemical data for this project. The reviews include an assessment of sample condition on receipt at the laboratory, compliance with holding times, instrument calibration, procedural blanks, laboratory control samples, standard reference materials, surrogate recoveries, matrix spike recoveries, and duplicate sample analyses. The full text of the data quality reviews and the complete chemical data reported by Manchester are available from the author on request.

Overall, the quality of the data is excellent. Some results required qualification for reasons outlined below.

Metals

Lead results were qualified as estimates (J flag) due to blank contamination and high recovery in a laboratory control sample. The lead concentrations reported here are likely biased high.

Pesticides

The reporting limits for endosulfan I, endrin ketone, and alpha-BHC were qualified as estimates due to low matrix spike recoveries. The reporting limits for heptachlor epoxide were qualified as estimates due to poor precision between control standards. None of these pesticides was detected in the samples.

PCBs

Low concentrations of PCBs were detected in some samples. The pattern most closely matched PCB-1248 but may have been weathered -1242. All PCB results were qualified as estimated concentrations below the reporting limit.

PAH

These data required no qualification.

Dioxins and Furans

Low concentrations of 2,3,7,8-tetrachlorodibenzofuran were detected in some samples. These results were qualified as estimates because the concentrations were below the lowest calibration standard.

Tributyltins

These data required no qualification.

Percent Lipids and Solids

These data required no qualification.

Analytical Precision

The precision of the data contained in this report can be gauged from results of duplicate analyses conducted for certain of the target chemicals (Table 4). Except for lead in littleneck clams from Crandall Spit, there was close agreement between duplicates.

Table 4. Analytical Precision on Split Samples (ug/Kg, wet weight)
[Detected compounds only]

Chemical	Analysis #1	Analysis #2
Dungeness Crab Muscle, Samish Island (Sample No. 218020)		
4,4'-DDE	0.24	0.26
PCB-1248	1.8 J	1.1 J
Littleneck Clam Soft Parts, Crandall Spit (Sample No. 228034)		
Lead	104	150
1,1'-Biphenyl	2.6 J	2.1 J
Phenanthrene	24	24 U
Fluoranthene	27	25
Pyrene	14	11
Benzo(a)anthracene	2.8	3.4
Chrysene	5.2	7.3
Oyster Soft Parts, Swinomish Channel (Sample No. 228031)		
Cadmium	1440	1480

U = not detected at or above reported value
J = estimated value

Results

Appendix B summarizes results from analysis of the shellfish samples, showing the reporting limits for each of the chemicals analyzed. The discussion and tables that follow are limited to those chemicals detected in one or more samples. The data are reported on a wet-weight basis in units of ug/Kg (parts per billion) or ng/Kg (parts per trillion).

Crab Muscle

The concentrations of metals and organic compounds detected in Dungeness crab muscle are shown in Table 5. Of the five metals analyzed, the highest concentrations were found for arsenic at 5,230 - 8,390 ug/Kg, followed by selenium at 496 - 692 ug/Kg, mercury at 41 - 75 ug/Kg, and lead at 11 - 33 ug/Kg. Cadmium was not detected at or above 25 ug/Kg in any of the muscle tissue samples (Appendix B). Metals concentrations in crabs from the Samish Island reference area were not substantially different from those in crabs from other sites, and for lead and mercury were slightly higher than samples from the March Point and Fidalgo Bay area.

Very few organic compounds were detected in crab muscle. The only pesticide identified was 4,4'-DDE, a breakdown product of DDT. Similar low concentrations of 0.19 - 0.25 ug/Kg DDE were detected in the March Point, Hat Island, and Samish Island crabs. DDE was not detected in the Fidalgo Bay crab sample, but the reporting limit (0.26 ug/Kg) was close to the levels seen elsewhere.

PCB-1248 was identified in the Hat and Samish Island crabs at 1.2 and 1.4 ug/Kg, respectively. As with DDE, the reporting limit for PCBs in the other muscle tissue samples from March Point and Fidalgo Bay was close to this level (2.1 - 2.6 ug/Kg). It is probably that PCBs and DDE were present in all crab muscle samples at roughly comparable concentrations.

Only 2,3,7,8-tetrachlorodibenzofuran (TCDF) was detected in the analysis for polychlorinated dioxins and -furans. The detections again occurred in the Hat and Samish Island samples, at 0.49 and 0.51 ng/Kg, respectively. The reporting limit for TCDF in Fidalgo Bay crabs was slightly lower at 0.39 ng/Kg, and for March Point slightly higher at 0.78 ng/Kg. The reporting limits for 2,3,7,8-tetrachlorodibenzo-p- dioxin (TCDD, "dioxin") were 0.40 - 0.80 ng/Kg; none was detected (Appendix B).

C-1 naphthalenes (methylnaphthalenes) were detected in the Samish Island crab sample at an estimated concentration of 4.9 ug/Kg. Reporting limits for C1-naphthalenes at the other locations sampled were 3.6 - 6.6 ug/Kg. No other PAH were detectable in any of the crab muscle samples.

The antifouling agent tributyltin was not detected in crab muscle at or above 2.6 ug/Kg (Appendix B).

Table 5. Chemicals Detected in Dungeness Crab Muscle (wet weight)
[Detections highlighted in **BOLD**]

Sampling Site	March Point	Fidalgo Bay	Hat Island	Samish Island
Collection Date	26-May-99	26-May-99	26-May-99	26-May-99
Sample Number	218023	218022	218021	218020
Metals (ug/Kg)				
Arsenic	**7350**	**5230**	**8390**	**5700**
Lead	**20** J	**11** J	**33 J**	**29** J
Selenium	**496**	**512**	**692**	**591**
Mercury	**56**	**41**	**75**	**70**
Pesticides (ug/Kg)				
4,4'-DDE	**0.22**	0.26 U	**0.19**	**0.25**
PCBs (ug/Kg)				
PCB-1248	2.1 U	2.6 U	**1.2** J	**1.4** J
Dioxins and Furans (ng/Kg)				
2,3,7,8-TCDF	0.78 U	0.39 U	**0.49** J	**0.51** J
PAH (ug/Kg)				
C1-Naphthalenes	3.8 U	3.6 U	6.6 U	**4.9** NJ
Percent Lipid	0.1	0.1	0.1	0.1
Percent Solids	18	17	17	17

U = not detected at or above reported value
J = estimated value
NJ = evidence the analyte is present; numerical result is an estimate

Clams and Oysters

The clam and oyster data are in Table 6. As in crab muscle, the metal present in the highest concentrations was arsenic at 1,360 - 2,600 ug/Kg. The relative abundance of the other metals analyzed differed from results for crab tissue, in that cadmium was readily detected at 211 - 1,460 ug/Kg, concentrations roughly similar to selenium, 470 - 750 ug/Kg. Lead concentrations, 43 - 128 ug/Kg, exceeded mercury, 11 - 26 ug/Kg, also in contrast to results for crab.

The highest lead concentrations occurred in the March Point and Crandall Spit littleneck clams, 128 and 127 ug/Kg, respectively, 2 to 3 times higher than in clams or oysters from other

Table 6. Chemicals Detected in Clams and Oysters (wet weight basis)
[Detections highlighted in **BOLD**]

Species	Littleneck Clams			Oysters		Butter Clams
Sampling Site	March Point	Crandall Spit	Person Road	Bayview	Swinomish Ch	Samish Island
Collection Date	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99
Sample Number	228033	228034	228032	228030	228031	228035
Metals (ug/Kg)						
Arsenic	**2040**	**2310**	**2520**	**1460**	**1360**	**2600**
Lead	**128** J	**127** J	**51** J	**55** J	**43** J	**46** J
Cadmium	**317**	**410**	**211**	**822**	**1460**	**366**
Selenium	**535**	**674**	**750**	**660**	**470**	**599**
Mercury	**15**	**26**	**21**	**21**	**22**	**11**
Butyltins (ug/Kg)						
Tributyltin chloride	1.8 U	2.1 U	1.8 U	**2.7**	**7.9**	2.2 U
Pesticides (ug/Kg)						
4,4-DDT	0.25 U	0.23 U	0.25 U	**0.54**	**0.20**	0.25 U
4,4'-DDE	0.25 U	0.23 U	0.25 U	**1.4**	**0.65**	0.25 U
4,4'-DDD	0.25 U	0.23 U	0.25 U	**0.30**	**0.27**	0.25 U
Dieldrin	0.25 U	0.23 U	0.25 U	**0.25**	0.26 U	0.25 U
Trans-Nonachlor	0.32 U	0.29 U	0.31 U	**0.18** J	**0.22** J	0.32 U
PCBs (ug/Kg)						
PCB-1248	**2.3** J	2.3 U	2.5 U	**2.7** J	**1.3** J	2.5 U
PCB-1254	2.5 U	2.3 U	2.5 U	2.5 U	**1.7** J	2.5 U
Dioxins & Furans (ng/Kg)						
2,3,7,8-TCDF	0.45 U	0.49 U	1.10 U	**0.69** J	**0.46** J	0.37 U
PAH (ug/Kg)						
1,1'-Biphenyl	1.8 U	**2.4** J	3.9 U	3.9 U	1.9 U	3.7 U
1-Methylnaphthalene	0.93 U	0.91 U	0.97 U	0.97 U	**3.6**	0.94 U
2-Methylnaphthalene	0.93 U	0.91 U	0.97 U	0.97 U	**11**	0.94 U
1,6,7-Trimethylnaphthalene	0.93 U	0.91 U	0.97 U	0.97 U	**4.2**	0.94 U
Phenanthrene	14 U	**24**	0.97 U	12 U	**19**	0.94 U
1-Methylphenanthrene	0.93 U	0.91 U	0.97 U	0.97 U	4.3 U	0.94 U
2-Methylphenanthrene	0.93 U	0.91 U	0.97 U	0.97 U	4.9 U	0.94 U
Fluoranthene	**7.0**	**26**	0.97 U	**9.0**	**29**	0.94 U
Pyrene	**6.1**	**12**	0.97 U	4.7 U	**17**	0.94 U
Benzo[a]anthracene	0.93 U	**3.1**	0.97 U	0.97 U	**4.2**	0.94 U
Chrysene	0.93 U	**6.2**	0.97 U	0.97 U	**8.7**	0.94 U
Benzo[e]pyrene	0.93 U	0.91 U	0.97 U	0.97 U	2.9 U	0.94 U
C1-Naphthalenes	6.4 U	3.6 U	3.9 U	**6.8** NJ	**15** NJ	3.7 U
C2-Naphthalenes	3.7 U	3.6 U	3.9 U	3.9 U	**12** NJ	3.7 U
C3-Naphthalenes	3.7 U	3.6 U	3.9 U	3.9 U	**12** NJ	3.7 U
Percent Lipid	0.3	0.3	0.3	0.9	1.0	0.4
Percent Solids	13	13	10	17	16	14

U = not detected at or above reported value
J = estimated value
NJ = evidence the analyte is present; numerical result is an estimate

locations. Cadmium concentrations in both of the oyster samples from Bayview State Park, 822 ug/Kg, and the Swinomish Channel, 1,460 ug/Kg, were high compared to clams.

Many more organic compounds were detected in clams and oysters than in crabs, particularly in oysters. No organic compounds were detectable in the reference area butter clam sample.

Tributyltin was only found in oysters. There was 2.7 ug/Kg in the Bayview State Park sample and 7.9 ug/Kg in the Swinomish Channel sample. Reporting limits for tributyltin in the clam samples ranged from 1.8 - 2.2 ug/Kg.

Pesticides detected in the Bayview State Park and Swinomish Channel oysters included DDT, its breakdown products DDE and DDD, dieldrin (Bayview only), and trans-nonachlor. Except for trans-nonachlor, concentrations were higher at Bayview, from 0.18 - 1.4 ug/Kg. No pesticides were detectable in littleneck clams collected to the north at Person Road or from March Point or Crandall Spit.

Similar levels of 1.3 - 2.7 ug/Kg PCB-1248 were found in March Point littlenecks and in the Bayview and Swinomish Channel oysters. PCB-1254 was also identified at 1.7 ug/Kg in the Swinomish Channel oysters. These concentrations are close to the reporting limits for samples at other sites.

TCDF was again the only dioxin/furan detected, and in oysters only. The Bayview sample had 0.69 ng/Kg TCDF and the Swinomish Channel sample had 0.46 ng/Kg. TCDD was not detected in any of the clam or oyster samples at or above 0.41 - 0.99 ng/Kg (Appendix B).

A range of PAH compounds was detectable in the clam and oyster samples, primarily in the Swinomish Channel oysters and, to a lesser extent, the Crandall Spit littleneck clams. The compounds present in the highest concentrations here were fluoranthene at 26 - 29 ug/Kg, phenanthrene at 19 - 24 ug/Kg, pyrene at 12 - 17 ug/Kg, and substituted naphthalenes at 12 - 15 ug/Kg (Swinomish Channel oysters only). No PAH were detectable in the Person Road or Samish Island clams.

Crab Hepatopancreas and Mussels

Crab hepatopancreas and mussel soft parts from March Point and the Samish Island reference area were analyzed for PAH only (Table 7). The hepatopancreas is a lipid-rich organ and the site of digestive enzyme production, absorption, and food storage in crabs.

The PAH results on crab hepatopancreas were nearly identical. The same three compounds – 2,6-dimethylnaphthalene, benzo[e]pyrene, and C1-naphthalenes – were detected in both the March Point and Samish Island samples at comparable concentrations of 6.9 - 18 ug/Kg.

There were substantial differences in the levels of PAH in the mussel samples from these two sites. Numerous PAH were quantified in the March Point mussels, ranging from 2.5 ug/Kg of 1-methynaphthalene to 32 ug/Kg of C1-chrysenes. No PAH were detectable in the Samish Island mussels.

Table 7. PAH Compounds Detected in Crab Hepatopancreas and Mussel Soft Parts (wet weight basis)
[Detections highlighted in **BOLD**]

Species - Tissue	Dungeness Crab - Hepatopancreas		Mussel - Soft Parts	
Sampling Site	March Point	Samish Island	March Point	Samish Island
Collection Date	26-May-99	26-May-99	1-Jun-99	1-Jun-99
Sample Number	218025	218024	228036	228037
PAH (ug/Kg)				
2,6-Dimethylnaphthalene	**8.0**	**10**	3.4 U	3.8 U
Phenanthrene	15 U	12 U	**15**	6.8 U
1-Methylphenanthrene	0.99 U	0.99 U	**2.5**	0.95 U
2-Methylphenanthrene	0.99 U	0.99 U	**4.2**	0.95 U
Fluoranthene	0.99 U	0.99 U	**10**	0.95 U
Pyrene	0.99 U	0.99 U	**6.6**	0.95 U
Chrysene	0.99 U	0.99 U	**12**	0.95 U
Benzo[e]pyrene	0.99 U	0.99 U	**8.0**	0.95 U
C1-Naphthalenes	**7.3** NJ	**6.9** NJ	**5.6** NJ	4.8 U
C2-Naphthalenes	**18** NJ	**13** NJ	3.4 U	3.8 U
C3-Naphthalenes	4.0 U	3.9 U	**5.6** NJ	3.8 U
C1-Phenanthrenes/Anthracenes	4.0 U	3.9 U	**15** NJ	6.6 U
C2-Phenanthrenes/Anthracenes	4.0 U	3.9 U	**2.8** NJ	3.8 U
C1-Chrysenes	4.0 U	3.9 U	**32** NJ	3.8 U
Percent Lipid	4.9	13	0.8	1.0
Percent Solids	25	18	11	14

U = not detected at or above reported value
NJ = evidence the analyte is present; numerical result is an estimate

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Discussion

Species and Site Differences

This study used composite sampling to obtain representative data on a wide range of chemicals. Because only one sample per site was analyzed for each species and because, for some sites, only a single composite was analyzed, it is not possible to make conclusions about differences between species or sampling sites with a high degree of confidence. However, a few qualitative patterns were evident and are described below.

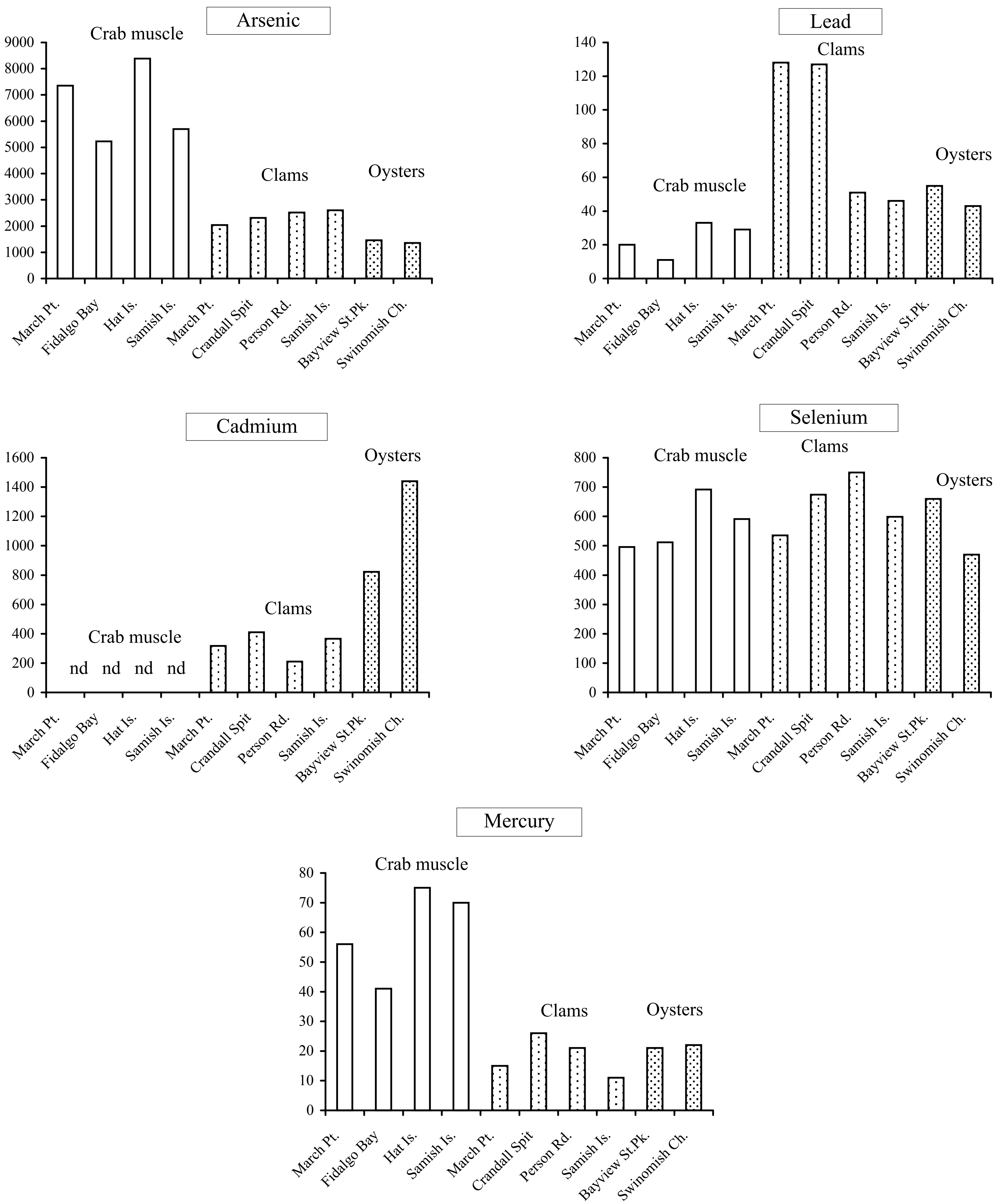
Figure 2 plots the data on metals concentrations in crab muscle, clams, and oysters. Crab muscle consistently had 2 to 6 times higher concentrations of arsenic and mercury than either clams or oysters. Concentrations of lead and especially cadmium were always greater in clams and oysters than in crabs. Oysters appeared to be strong accumulators of cadmium, with levels 2 to 7 times higher than clams. These same species differences in metals accumulation can be seen in other studies of Puget Sound shellfish (see Tables 8 and 9).

Allowing for the tendency for oysters to accumulate cadmium, the data show little evidence of differences in metals levels between sampling sites. Lead may be an exception, in that littleneck clams at both Crandall Spit and March Point had over twice the concentrations found in littleneck clams on the east shore of Padilla Bay, or in crabs, oysters, and clams from other areas.

Results for selected organic compounds detected in the shellfish samples are plotted in Figure 3. In this figure, total DDT refers to DDT+DDE+DDD and total PCBs to PCB-1248 + PCB-1254, where the latter was detected. The individual PAH compounds detected were summed to give Sum PAH. As previously noted, a number of chemicals were near the reporting limit, so this figure overemphasizes differences between samples.

Many of the organic compounds analyzed were not quantified consistently enough to draw strong conclusions about differences between species. Tributyltin was only detected in oysters. Clams had little or no DDT compounds, PCBs, or TCDF detected compared to crabs and oysters. Crab muscle had almost no PAH detectable compared to the other species analyzed, although several PAH were quantified in crab hepatopancreas.

Allowing for analytical variability (Table 4), several between-site differences are suggested by these results. Tributyltin concentrations are clearly elevated in Swinomish Channel oysters, while the levels of DDT compounds stand out in the Bayview State Park and Swinomish Channel oysters. Other pesticides, dieldrin and trans-nonachlor, were also detected only at these two sites. Three sites had much higher PAH concentrations than other locations: Crandall Spit (littleneck clams), Swinomish Channel (oysters), and March Point (mussels, but not littleneck clams).



nd = not detected

Figure 2. Metals Concentrations in Shellfish Samples (ug/Kg, wet weight)

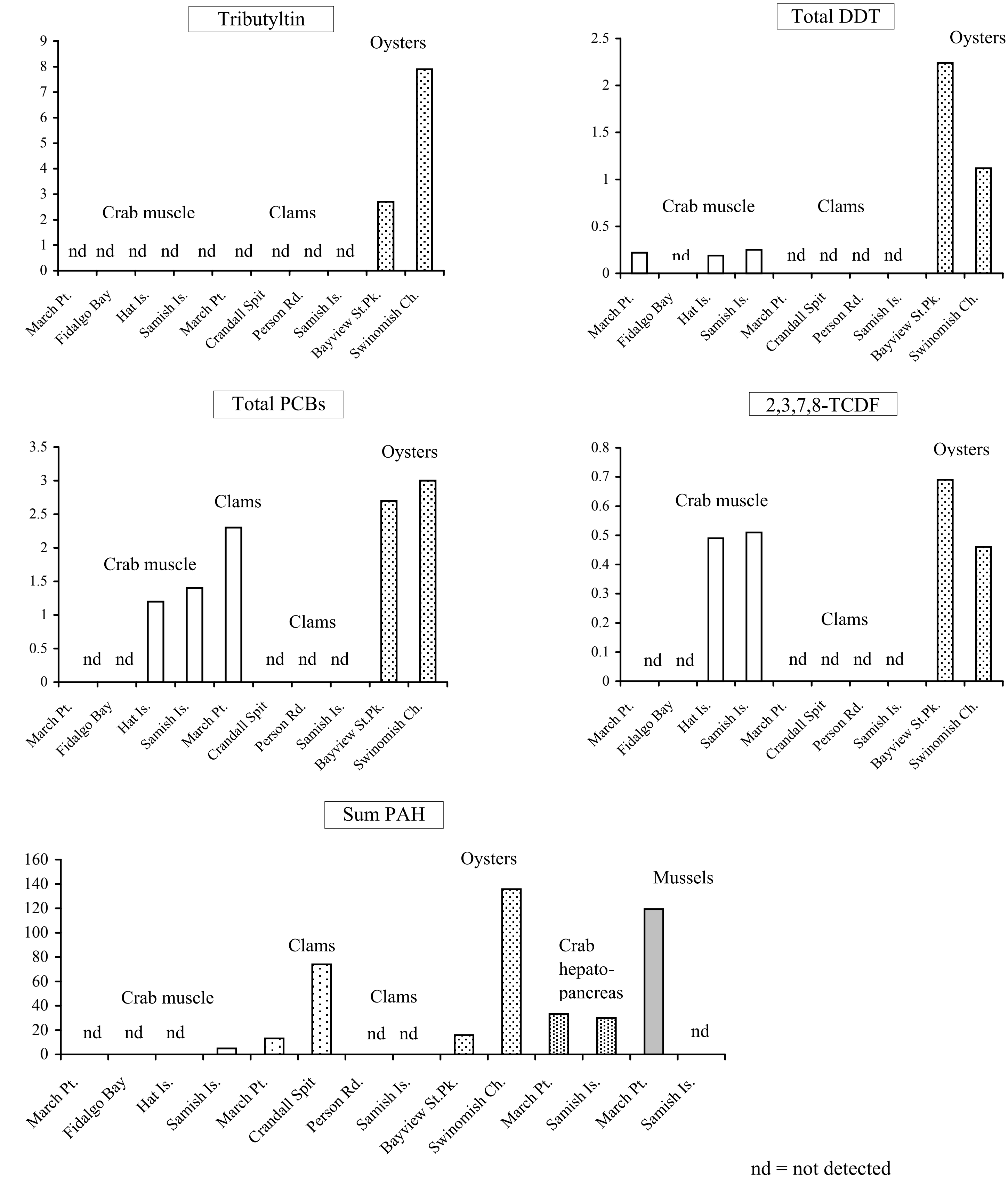


Figure 3. Selected Organic Compounds in Shellfish Samples (ug/Kg weight, except ng/Kg for TCDF)

Figure 4 plots the same data on a lipid-weight, rather than wet-weight, basis (wet-weight concentration divided by the decimal fraction of percent lipids). Because the compounds in question are preferentially soluble in lipid (fat), normalizing the data to lipid serves to remove between-sample differences that may be due solely to lipid content.

The normalized data bring the DDT levels in Bayview and Swinomish Channel oysters more in line with crabs from other parts of the study area, but tributyltin still stands out in the Swinomish Channel oysters. PCBs and TCDF in crab muscle appear elevated at Hat and Samish Islands when normalized. These two areas, however, are not near sources and the apparent elevations likely result from the measurements being made at the limits of detection.

For PAH, the normalized data show the same pattern as the wet-weight data. This supports the conclusion that Crandall Spit, March Point, and the Swinomish Channel have a higher level of PAH contamination than the other sites.

Sources

Except perhaps for lead, results of this study fail to point to significant sources affecting metals levels in the study area. Potential sources of lead at Crandall Spit and March Point, where elevations were found in clams, include wastewater treatment plant discharge, stormwater, road runoff, and refinery effluents. As noted earlier in this report, it is likely that the lead data are biased high. Even so, the concentrations at Crandall Spit and March Point are not particularly high compared to clams from other parts of Puget Sound (see p. 26).

Given the heavy concentration of marinas, boatyards, and vessel traffic in the Swinomish Channel, some elevation in tributyltin levels in oysters is to be expected. The tributyltin concentration in the Bayview State Park oyster sample was near the limits of detection in samples from other areas, so this finding should not be taken to indicate the presence of significant sources near the park.

The pesticides and PCBs detected in shellfish from the study area were banned in the 1970s or early 1980s. DDT compounds and PCBs are the most frequently reported organic chemicals in Puget Sound biological samples. The levels found in Padilla and Fidalgo Bay shellfish likely reflect present day background. The greater number of pesticides detectable in oysters from the Bayview and Swinomish Channel sites may be due to their closer proximity to agricultural sources, but again the concentrations are low.

Dioxins and furans, including TCDF, have no commercial uses, but are unintended byproducts formed during combustion of organic matter in the presence of chlorine, incineration of municipal and hospital wastes, and chlorine bleaching of wood pulp (Yake et al., 1998). Because of cost, they are infrequently analyzed in Puget Sound biota. In those instances where they have been analyzed, TCDF is one of the most frequently reported compounds (PTI, 1991a; START, 1999; Johnson, 1988). As shown below, the levels found in Padilla and Fidalgo Bay shellfish are extremely low.

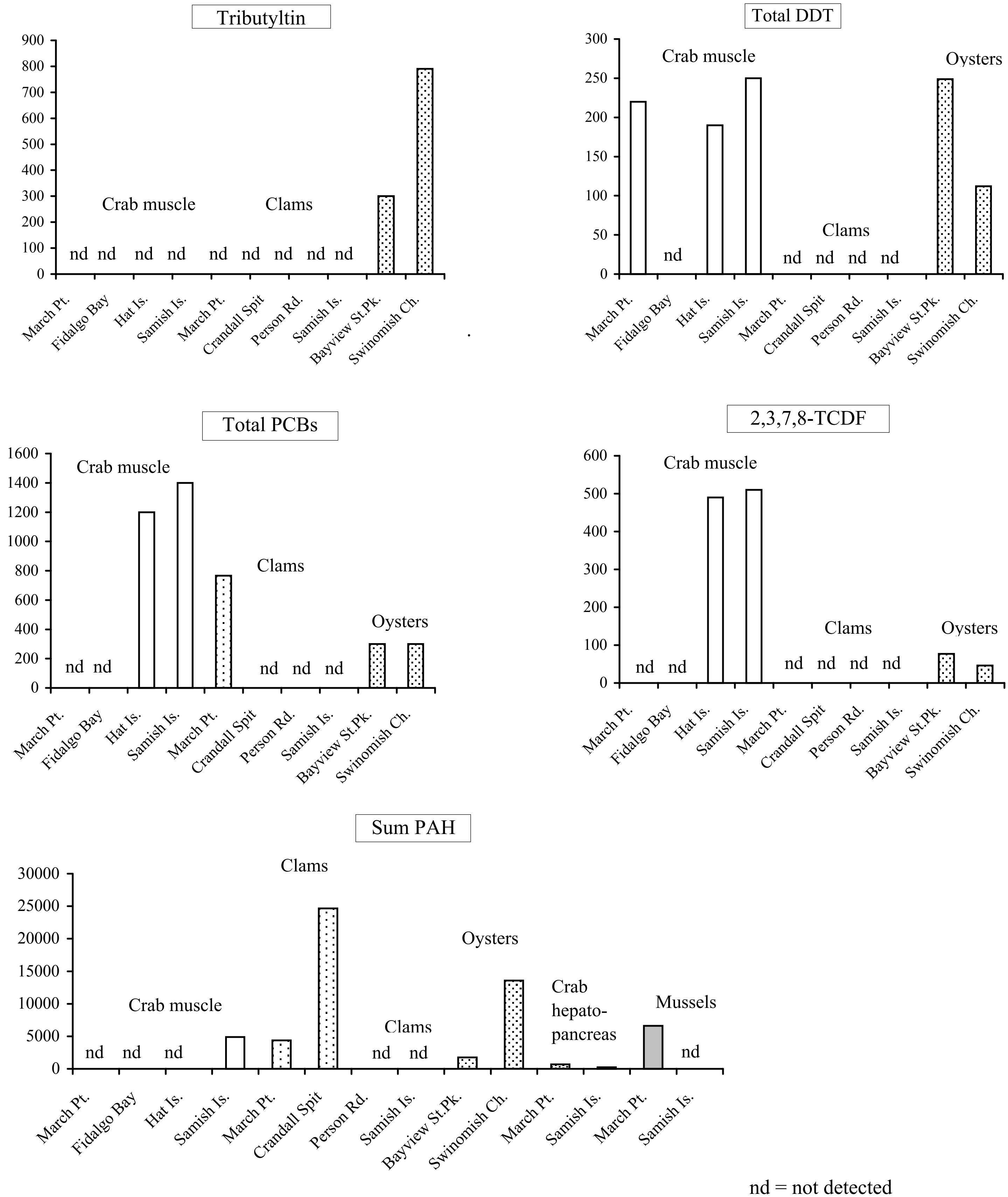


Figure 4. Selected Organic Compounds in Shellfish Samples (ug/Kg lipid, except ng/Kg for TCDF)

PAH compounds are present in petroleum and formed during combustion of fossil fuels. Petroleum sources have a higher percentage of alkyl-substituted low molecular weight PAH relative to the parent compound (e.g., methylnaphthalenes vs. naphthalene) compared to combustion sources (Lake et al., 1979). Only in a few samples were sufficient PAH quantified to see patterns. Petroleum-derived PAH are indicated in the Swinomish Channel oyster sample and in the Hat and Samish Island crab samples. Combustion PAH dominate in the Crandall Spit littlenecks and March Point mussels.

Finally, the results for crabs, clams, and mussels from Samish Island show an absence of any detectable influence from nearby sources of chemical contamination, and illustrate its appropriateness for use as a reference area.

Comparison to Other Studies

A literature review was conducted to locate other recent studies on chemical contaminants in shellfish from the Padilla Bay area and greater Puget Sound. Only limited data were available on Padilla or Fidalgo Bay.

The U.S. Fish and Wildlife Service (1994) analyzed metals and PAH in littleneck clams, softshell clams, and oysters from six sites in Padilla Bay and three sites in Fidalgo Bay in 1988. They concluded that the occurrence of "elevated chemical residues were few and localized." Some elevations were seen for PAH in softshell clams near March Point.

WDOH analyzed EPA priority metal and organic pollutants in littleneck clams collected from Crandall Spit and Samish Bay in 1992 and 1993, as part of PSAMP (Patrick, 1996). Results were comparable to clams from other WDOH monitoring sites in Puget Sound.

Ecology analyzed bioaccumulative pesticides and PCBs in a composite mussel tissue sample collected from the east shore of Padilla Bay in 1995 (Johnson and Davis, 1996). Trace amounts (< 1 ug/Kg) of six pesticides or their breakdown products were detected: DDT, DDE, DDD, endosulphan, alpha BHC, and hexachlorobenzene.

This sample also contained an estimated 2 ug/Kg of PCB-1254. Although low compared to tissue samples from many other parts of Puget Sound, the PCB concentration exceeded EPA's National Toxics Rule (NTR) human health criterion of 1.4 ug/Kg. This finding is the reason for Padilla Bay currently being on Ecology's 303(d) list, *Impaired and Threatened Waterbodies Requiring Additional Pollution Controls*. EPA has since revised the NTR PCB criterion to 5.3 ug/Kg (EPA, 1999).

During 1991, EPA conducted a survey of polychlorinated dioxins and -furans in Puget Sound crabs (PTI, 1991a). Muscle and hepatopancreas were analyzed from specimens collected in nine embayments near potential sources (pulp mills, oil refineries, wastewater treatment plants, wood treaters, marinas, shipyards, and agriculture). One muscle and one hepatopancreas sample were analyzed from Dungeness crab collected near the March Point refineries. TCDF and several higher chlorinated dioxins and -furans were detected in the muscle sample. The concentrations in both muscle and hepatopancreas were comparable to crabs from Dungeness Bay, the study's reference area.

Data from the above-mentioned studies and results from the present investigation are summarized and compared to similar data for other parts of Puget Sound in Tables 8 through 12. Examination of these data shows no instance where the concentrations of metals or organic compounds detected in shellfish from Padilla or Fidalgo Bay are substantially elevated compared to the same species analyzed elsewhere. For example, embayments that have had large sources of mercury (Bellingham Bay), lead (Sinclair Inlet, Duwamish River), tributyltin (Duwamish River), PCBs (Elliott Bay, Duwamish River, Commencement Bay), and dioxins/furans (Everett Harbor) have tissue concentrations in shellfish at least twice as high and often an order of magnitude higher. For most chemicals, the concentrations measured in shellfish from the Padilla/Fidalgo Bay area appear to be at present day background for Puget Sound.

No PAH data were located that were comparable to the extensive, low-level analysis conducted for the present study. However, the PAH concentrations measured in Padilla and Fidalgo Bay shellfish are, for the most part, clearly very low (Table 12).

Table 8. Metals Concentrations Reported in Studies on Puget Sound Dungeness Crab Muscle (ug/Kg, wet weight) [median values]

Location	Reference	N =	Arsenic	Lead	Cadmium	Selenium	Mercury
Padilla/Fidalgo Bay	**present study**	**3**	**7350**	**20**	**<25**	**512**	**56**
Samish Island*	**" "**	**1**	**5700**	**29**	**<25**	**591**	**70**
Bellingham Bay	Cubbage (1991)	8	3720	130	3	na	100
Everett Harbor	PTI (1988)	10	na	na	na	na	70
Port Susan*	" "	1	na	na	na	na	70
Elliott Bay	King Co. (1999)	3	11100	155	85	na	61
Duwamish River	" " "	2	9945	243	17	na	100
Port Susan*	" " "	3	3250	31	27	na	66
Port Angeles	START (1999)	3	5850	17	212	743	44
Dungeness Bay*	" "	1	22000	<10	107	1380	89

* reference area
na = not analyzed

Table 9. Metals Concentrations Reported in Studies on Puget Sound Clams and Oysters (ug/Kg, wet weight) [median values except as noted]

Location	Reference	Species	N =	Arsenic	Lead	Cadmium	Selenium	Mercury
Padilla/Fidalgo Bay	**present study**	**Littleneck**	**3**	**2310**	**127**	**317**	**674**	**21**
Samish Island*	**" "**	**Butter**	**1**	**2600**	**46**	**366**	**599**	**11**
Padilla/Fidalgo Bay	USFWS (1994)	Littleneck	5	3360	106	210	770	36
Samish Island*	" "	Littleneck	1	4640	110	680	640	20
March Point	Patrick (1996)[a]	Littleneck	3	1830	40	210	na	10
Samish Bay	" "	"	3	3070	60	330	na	30
Puget Sound	" "	"	57	1930	50	240	na	10
Bellingham Bay	Cubbage (1991)	Littleneck	4	1620	90	216	na	10
King County Beaches	Stark (1999)	Butter	10	3200	130	61	340	9
Sinclair/Dyes Inlets	Cubbage	Littleneck	8	2450	260	440	na	30
Dyes Inlet	Johnson (1998)	Littleneck	12	3635	na	na	na	43
Hood Canal*	" "	"	2	2560	na	na	na	<5
South Puget Sound	Norton (1988)	Littleneck	4	1350	na	340	na	11
" " "	" "	Butter	4		na	110	na	10
Bayview St. Park	**present study**	**Oyster**	**1**	**1460**	**55**	**822**	**660**	**21**
Swinomish Channel	**" "**	**"**	**1**	**1360**	**43**	**1460**	**470**	**22**
Padilla Bay	USFWS (1994)	Oyster	1	1160	120	960	330	44

*reference area

[a]1993 data only, mean values

na = not analyzed

Table 10. Concentrations of Organic Compounds Reported in Studies on Puget Sound Dungeness Crab Muscle (ug/Kg, wet weight, except ng/Kg for TCDF and TEQs)
[median values, except as noted]

Location	Reference	N =	TBT	T-DDT	T-PCBs	TCDF	Dioxin TEQs*
Padilla/Fidalgo Bay	**present study**	**3**	**<1.9**	**0.22**	**1.2[a]**	**0.49**	**0.049**
Samish Island**	**" "**	**1**	**<2.6**	**0.25**	**1.4**	**0.51**	**0.051**
Bellingham Bay	Cubbage (1991)	8	na	<2	<20	na	na
March Point	PTI (1991)	1	na	na	na	0.95	0.18
Bellingham Bay	" "	1	na	na	na	0.96	0.13
Everett Harbor	" "	1	na	na	na	2.7	0.31
West Point	" "	1	na	na	na	1.2	0.12
Dungeness Bay **	" "	1	na	na	na	<0.10	0.023
Port Angeles	START (1999)	3	na	0.83	13	0.58	0.06
Dungeness Bay**	" "	1	na	0.87	1.5	<0.26	0
Everett Harbor	PTI (1988)	10	na	<1	8	na	na
Port Susan**	" "	1	na	<1	5	na	na
Elliott Bay	King Co. (1999)	3	72	na	113	na	na
Duwamish River	" " "	2	64	na	156	na	na
Port Susan**	" " "	3	2.3	na	<5.3	na	na
Port Angeles	START (1999)	3	na	0.83	13	0.58	0.06
Dungeness Bay**	" "	1	na	0.87	1.5	<0.26	0
Elliott Bay	Ylitalo (1999)	10	na	na	120[b]	na	na
Commencment Bay	" "	10	na	na	43[b]	na	na
Nisqually Reach	" "	10	na	na	8.6[b]	na	na
Useless Bay**	" "	10	na	na	5.7[b]	na	na

*sum of dioxin toxic equivalents (TEQs) for all detected polychlorinated dioxins and -furans
**reference area
na = not analyzed
[a] maximum value
[b] mean value

Table 11. Concentrations of Organic Compounds Reported in Studies on Puget Sound Clams and Oysters (ug/Kg, wet weight; except ng/Kg for TCDF and TEQs) [median values]

Location	Reference	Species	N =	TBT	T-DDT	T-PCBs	TCDF
Padilla/Fidalgo Bay	**present study**	**Littleneck**	**3**	**<1.8**	**<0.23**	**2.3**[a]	**<0.49**
Samish Island*	**" "**	**Butter**	**1**	**<2.2**	**<0.25**	**<0.25**	**<0.37**
Padilla Bay/Swinomish Channel	**" "**	**Oyster**	**2**	**5.3**	**1.7**	**2.8**	**0.58**
March Point	Patrick (1996)[b]	Littleneck	3	na	<0.7	<8	na
Samish Bay*	" "	"	3	na	<0.7	<8	na
Puget Sound	" "	"	57	na	<0.7	<8	na
Bellingham Bay	Cubbage (1991)	Littleneck	4	na	<4	<20	na
King County Beaches	Stark (1999)	Butter	10	na	<1.3	<13	na
Sinclair/Dyes Inlets	Cubbage (1992)	Littleneck	8	na	<2	<20	na
South Puget Sound	Norton (1988)	Littleneck	4	na	<4	<40	na
"	"	Butter	4	na	<4	<40	na

* reference area
[a] maximum value
[b] 1993 data only
na = not analyzed

Table 12. Detection Frequency and Concentrations of PAH Compounds Reported in Other Studies on Puget Sound Shellfish (ug/Kg, wet weight)

Location	Reference	Species	N =	Detection Frequency	Concentrations Detected
Padilla/Fidalgo Bay	**present study**	**Crab muscle**	**3**	**1/4**	**4.9**
" "	**" "**	**Crab hepatopancreas**	**2**	**2/2**	**1.8 - 26**
" "	**" "**	**Clams (two sp.)**	**4**	**2/4**	**1.8 - 27**
" "	**" "**	**Oysters**	**2**	**2/2**	**3.6 - 29**
" "	**" "**	**Mussels**	**2**	**1/2**	**2.5 - 32**
Padilla/Fidalgo Bay	USFWS (1994)	Clams (two sp.)	10	10/10	10 - 40
" "	" "	Oyster	1	1/1	10 - 30
March Point	Patrick (1996)[a]	Littleneck clams	3	0/3	nd
Puget Sound	" "	" "	57	1/57	33 - 132
Bellingham Bay	Cubbage (1991)	Littleneck clams	4	4/4	3 - 10
King County Beaches	Stark (1999)	Butter clams	10	0/10	nd
Elliott Bay/Duwamish R.	King Co. (1999)	Crab muscle	5	2/5	11 -226
Port Susan*	" " "	" "	3	0/3	nd
Sinclair/Dyes Inlets	Cubbage	Littleneck clams	8	8/8	4 - 20
South Puget Sound	Norton (1988)	Clams (various sp.)	8	0/8	nd

*reference area
[a]1993 data only
nd = not detected

Human Health Implications

The WDOH Office of Environmental Health Assessments (OEHA) reviewed the shellfish data obtained during this study, to determine if the chemical concentrations detected indicate the presence of a human health concern and thus warrant more intensive chemical and species-specific sampling and analysis (Patrick, 2000). Their review and conclusions are summarized below. Appendix C has the complete text of the WDOH report.

OEHA conducted a screening evaluation based on human health toxicity endpoints, using the approach outlined in EPA (1995). Screening chemical concentration values were calculated for 23 chemicals that had human health toxicity criteria, of the 30 chemicals detected in the shellfish samples. The screening values reflect a daily chemical intake level which is unlikely to result in any adverse human health impacts over an individual's lifetime exposure.

To make the screening values applicable to high shellfish consumers such as tribal members, species-specific consumption rates from the Tulalip Tribe were used (Toy et al., 1996). Ninetieth percentile shellfish consumption rates for Tulalip men were assumed for all chemicals except mercury, where the 90^{th} percentile rate for Tulalip women of childbearing age (18 - 49) was used. The most sensitive toxicological effect for mercury occurs in prenatally exposed children. Due to a lack of consumption information, the crab hepatopancreas PAH data were not evaluated with regard to human health implications.

Appendix C shows each of the screening values OEHA calculated for crab muscle, clams, oysters, and mussels. Two chemicals were identified as being of potential human health concern. Arsenic concentrations detected in all shellfish samples except mussels, where it was not analyzed, exceeded calculated screening values. The reported concentration of TCDF in the Dungeness crab sample from Samish Island was also at the calculated screening value based on a dioxin toxicity equivalency (TEQ) factor of 0.1 for this compound.

Although the arsenic concentrations were generally comparable to shellfish from other parts of Puget Sound, lack of data on the chemical form of arsenic lead to uncertainty by OEHA regarding conclusions about the presence of health risk due to shellfish arsenic concentrations. Arsenic toxicity is primarily due to inorganic arsenic. The OEHA evaluation assumed 10 percent of the total arsenic concentration was in an inorganic form.

The issue of arsenic in Puget Sound shellfish is problematic, and one of ongoing research and discussion at OEHA. To help reduce uncertainties associated with assessing the human health implications of arsenic in shellfish, OEHA recommended that further sampling and analysis of shellfish for arsenic be conducted, including measurement of both total arsenic and speciated arsenic during different seasons of the year.

TCDF concentrations were near the analytical detection limit for this compound and therefore may reflect the ambient concentration of this compound in Dungeness crab within Puget Sound. While scientists and health professionals worldwide acknowledge that exposure to dioxins and furans should be minimized, there is currently no agreement on what level of exposure constitutes a tolerable daily intake. Given the limitations of the toxicological understanding of this compound, and the background level of reported concentrations, additional shellfish collection and analysis for dioxins and furans was not recommended by OEHA.

Conclusions and Recommendations

Conclusions

Results of this study show that shellfish in the Padilla/Fidalgo Bay area have a very low level of chemical contamination. In most instances, the concentrations detected appear to be at background levels for Puget Sound. The influence of urban/industrial sources is detectable at some locations, but barely so.

Most of the chemicals analyzed were at concentrations that should not pose a significant human health threat for tribal or recreational consumption of shellfish. Arsenic and TCDF exceeded human health screening levels, but the significance of this finding is uncertain.

Recommendations

- Conduct further sampling and analysis for arsenic in frequently consumed shellfish from the study area, including samples representing different seasons of the year. Use state-of-the-art analytical techniques to measure total arsenic and inorganic arsenic species.

- Remove Padilla Bay from the 303(d) list for exceeding the National Toxics Rule human health criteria for PCBs.

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Appendices

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Appendix A

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Appendix A. Samples Analyzed by Ecology for Chemical Screening of Padilla and Fidalgo Bay Shellfish

Location	Description	Latitude	Longitude	Date	Species	N =	Size Range (inches)
Swinomish Channel	West bank near red can #12, south end of riprap	48 25 36	122 29 59	1-Jun-99	Japanese Oyster	20	3 1/2 - 6
Bayview State Park	Off north end of parking lot	48 29 03	122 28 46	1-Jun-99	Japanese Oyster	20	2 1/2 - 6 1/4
Person Road	East side of Padilla Bay, off Person Road	48 30 27	122 29 01	1-Jun-99	Littleneck Clam	50	1 1/2 - 2
March Point	Due east of tank #216	48 29 49	122 33 16	1-Jun-99	Littleneck Clam	50	1 1/2 - 2 1/4
"	1/3 mile east of Tesoro wharf	48 30 26	122 33 40	26-May-99	Dungeness Crab	5	6 1/2 - 6 5/8
"	Concrete pilings, north end Tesoro pier, east corner	48 30 31	122 33 59	1-Jun-99	Bay Mussel	50	1 1/2 - 2 3/16
Crandall Spit	South side of spit about 200 yards west of oil pipeline	48 29 27	122 34 40	1-Jun-99	Littleneck Clam	50	1 1/2 - 2 3/8
Fidalgo Bay	Inner bay north of BNRR bridge	48 29 32	122 35 04	26-May-99	Dungeness Crab	5	6 1/4 - 7
Hat Island	Between Hat and Saddlebag Islands	48 32 05	122 33 11	26-May-99	Dungeness Crab	5	6 1/4 - 7

Appendix A. (continued)

Location	Description	Latitude	Longitude	Date	Species	N =	Size Range (inches)
Samish Island	Camp Kirby, southwest tip of Samish Island	48 34 26	122 33 24	1-Jun-99	Butter Clam	50	1 1/2 - 2 1/2
"	Northeast side of Samish Island, inside William Pt.	48 35 03	122 32 59	1-Jun-99	Bay Mussel	50	1 3/8 - 2
"	1/4 mile inside William Pt.	48 35 01	122 32 54	26-May-99	Dungeness Crab	5	6 1/2 - 6 7/8

Appendix B

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Appendix B. Ecology Data from Chemical Screening of Shellfish from Padilla Bay and Vicinity (wet weight basis)

Species	Oyster		Littleneck Clam			Butter Clam
Tissue	Soft Parts	Soft Parts	Soft Parts	Soft Parts	Soft Parts	Soft Parts
Sampling Site	Bayview St. Park	Swinomish Ch.	Person Road	March Point	Crandall Spit	Samish Island
Collection Date	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99
Sample Number	228030	228031	228032	228033	228034	228035
Metals (ug/Kg)						
Arsenic	**1460**	**1360**	**2520**	**2040**	**2310**	**2600**
Lead	**55** J	**43** J	**51** J	**128** J	**127** J	**46** J
Cadmium	**822**	**1460**	**211**	**317**	**410**	**366**
Selenium	**660**	**470**	**750**	**535**	**674**	**599**
Mercury	**21**	**22**	**21**	**15**	**26**	**11**
Pesticides (ug/Kg)						
Alpha-BHC	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Beta-BHC	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Gamma-BHC (Lindane)	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Delta-BHC	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Heptachlor	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Aldrin	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Heptachlor Epoxide	0.25 UJ	0.26 UJ	0.25 UJ	0.25 UJ	0.23 U	0.25 UJ
Trans-Chlordane (Gamma)	0.63 U	0.64 U	0.63 U	0.63 U	0.58 U	0.64 U
Endosulfan I	0.25 UJ	0.26 UJ	0.25 UJ	0.25 UJ	0.23 U	0.25 UJ
Dieldrin	**0.25**	**0.65**	0.25 U	0.25 U	0.23 U	0.25 U
4,4'-DDE	**1.4**	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Endrin	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Endosulfan II	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
4,4'-DDD	**0.30**	**0.27**	0.25 U	0.25 U	0.23 U	0.25 U
Endrin Aldehyde	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Endosulfan Sulfate	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
4,4'-DDT	**0.54**	**0.20**	0.25 U	0.25 U	0.23 U	0.25 U
Endrin Ketone	0.25 UJ	0.26 UJ	0.25 UJ	0.25 UJ	0.23 U	0.25 UJ
Methoxychlor	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Alpha-Chlordene	0.32 UJ	0.32 UJ	0.31 UJ	0.32 UJ	0.29 U	0.32 UJ
Gamma-Chlordene	0.32 UJ	0.32 UJ	0.31 UJ	0.32 UJ	0.29 U	0.32 UJ
Oxychlordane	0.32 U	0.32 U	0.31 U	0.32 U	0.29 U	0.32 U
DDMU	0.32 U	0.32 U	0.31 U	0.32 U	0.29 U	0.32 U
Cis-Chlordane (Alpha)	0.25 U	0.26 U	0.25 U	0.25 U	0.23 U	0.25 U
Cis-Nonachlor	0.32 U	0.32 U	0.31 U	0.32 U	0.29 U	0.32 U
Kelthane (Dicofol)	1.3 U	1.3 U	1.3 U	1.3 U	1.2 U	1.3 U
2,4'-DDE	0.32 U	0.32 U	0.31 U	0.32 U	0.29 U	0.32 U
Trans-Nonachlor	**0.18**	**0.45**	0.31 U	0.32 U	0.29 U	0.32 U
2,4'-DDD	0.51 U	0.21 U	0.31 U	0.32 U	0.29 U	0.32 U
2,4'-DDT	0.45 U	0.41 U	0.31 U	0.32 U	0.29 U	0.32 U
Mirex	0.32 U	0.32 U	0.31 U	0.32 U	0.29 U	0.32 U
Toxaphene	13 U	13 U	13 U	13 U	12 U	13 U
4,4'-Dichlorobenzophenone	1.3 U	1.3 U	1.3 U	1.3 U	1.2 U	1.3 U
Hexachlorobenzene	0.32 U	0.32 U	0.31 U	0.32 U	0.29 U	0.32 U
Pentachloroanisole	0.32 U	0.32 U	0.31 U	0.32 U	0.29 U	0.32 U
Tetradifon (Tedion)	1.3 U	1.3 U	1.3 U	1.3 U	1.2 U	1.3 U
DCPA (dacthal)	0.63 U	0.64 U	0.63 U	0.63 U	0.58 U	0.64 U
Diazinon	63 U	64 U	63 U	63 U	58 U	64 U
Ethion	25 U	26 U	25 U	25 U	23 U	25 U
Parathion	1.3 U	1.3 U	1.3 U	1.3 U	1.2 U	1.3 U
Methyl Parathion	1.3 U	1.3 U	1.3 U	1.3 U	1.2 U	1.3 U
Treflan (Trifluralin)	1.3 U	1.3 U	1.3 U	1.3 U	1.2 U	1.3 U
Chlorpyriphos	0.63 U	0.64 U	0.63 U	0.63 U	0.58 U	0.64 U
Oxadiazon	1.3 U	1.3 U	1.3 U	1.3 U	1.2 U	1.3 U

Appendix B. (continued)

Species	Oyster		Littleneck Clam			Butter Clam
Tissue	Soft Parts	Soft Parts	Soft Parts	Soft Parts	Soft Parts	Soft Parts
Sampling Site	Bayview St. Park	Swinomish Ch.	Person Road	March Point	Crandall Spit	Samish Island
Collection Date	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99
Sample Number	228030	228031	228032	228033	228034	228035
PCBs (ug/Kg)						
PCB - 1260	2.5 U	2.6 U	2.5 U	2.5 U	2.3 U	0.25 U
PCB - 1254	2.5 U	**1.7** J	2.5 U	2.5 U	2.3 U	0.25 U
PCB - 1221	2.5 U	2.6 U	2.5 U	2.5 U	2.3 U	0.25 U
PCB - 1232	2.5 U	2.6 U	2.5 U	2.5 U	2.3 U	0.25 U
PCB - 1248	**2.7** J	**1.3** J	2.5 U	**2.3** J	2.3 U	0.25 U
PCB - 1016	2.5 U	2.6 U	2.5 U	2.5 U	2.3 U	0.25 U
PCB - 1242	2.5 U	2.6 U	2.5 U	2.5 U	2.3 U	0.25 U
Dioxins & Furans (ng/Kg)						
2378-TCDF	**0.69** J	**0.46** J	1.10 U	0.45 U	0.49 U	0.37 U
2378-TCDD	0.41 U	0.57 U	0.80 U	0.55 U	0.99 U	0.70 U
12378-PeCDF	0.63 U	1.10 U	0.68 U	0.62 U	0.83 U	0.48 U
23478-PeCDF	0.78 U	1.60 U	0.61 U	0.34 U	0.36 U	0.39 U
12378-PeCDD	0.96 U	1.20 U	1.00 U	0.84 U	1.30 U	0.55 U
123478-HxCDF	0.87 U	1.40 U	1.10 U	0.56 U	0.94 U	0.64 U
123678-HxCDF	0.61 U	1.00 U	1.20 U	0.67 U	1.20 U	0.31 U
234678-HxCDF	0.81 U	2.10 U	3.60 U	0.36 UJ	0.81 U	0.63 U
123789-HxCDF	1.20 U	3.00 U	1.10 U	0.74 U	1.10 U	0.69 U
123478-HxCDD	0.57 U	2.00 U	1.40 U	0.52 U	0.76 U	0.50 U
123678-HxCDD	0.74 U	0.76 U	1.70 U	0.53 U	0.38 U	0.36 U
123789-HxCDD	0.53 U	0.90 U	0.82 U	0.29 U	0.75 U	0.36 U
1234678-HpCDF	0.97 U	2.10 U	2.00 U	0.45 U	1.30 U	1.30 U
1234789-HpCDF	1.50 U	3.50 U	1.90 U	1.60 U	0.92 U	1.20 U
12234678-HpCDD	1.00 U	2.90 U	1.90 U	1.10 U	1.60 UJ	1.80 U
OCDF	0.87 U	1.40 U	1.50 U	0.98 U	1.60 U	2.50 U
OCDD	1.90 U	2.90 U	10 U	2.60 UJ	7.20 UJ	6.50 UJ
PAH (ug/Kg)						
Naphthalene	0.97 U	4.9 U	0.97 U	0.93 U	0.91 U	0.94 U
2-Methylnaphthalene	0.97 U	**11**	0.97 U	0.93 U	0.91 U	0.94 U
1-Methylnaphthalene	0.97 U	**3.6**	0.97 U	0.93 U	0.91 U	0.94 U
1,1'-Biphenyl	3.9 U	1.9 U	3.9 U	1.8 U	**2.4** J	3.7 U
2-Chloronaphthalene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
2,6-Dimethylnaphthalene	3.9 U	4.1 U	3.9 U	3.7 U	3.6 U	3.7 U
Acenaphthylene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Acenaphthene	0.97 U	3.7 U	0.97 U	0.93 U	0.91 U	0.94 U
Dibenzofuran	0.97 U	4.1 U	0.97 U	0.93 U	0.91 U	0.94 U
1,6,7-Trimethylnaphthalene	0.97 U	**4.2**	0.97 U	0.93 U	0.91 U	0.94 U
Fluorene	0.97 U	2.5 U	0.97 U	0.93 U	0.91 U	0.94 U
9H-Fluorene, 1-methyl-	0.97 U	4.1 U	0.97 U	0.93 U	0.91 U	0.94 U
Dibenzothiophene	0.97 U	7.6 U	0.97 U	0.93 U	0.91 U	0.94 U
Phenanthrene	12 U	**19**	0.97 U	14 U	**24**	0.94 U
Anthracene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
2-Methylphenanthrene	0.97 U	4.9 U	0.97 U	0.93 U	0.91 U	0.94 U
1-Methylphenanthrene	0.97 U	4.3 U	0.97 U	0.93 U	0.91 U	0.94 U
4,6-Dimethyldibenzothiophene	0.97 U	3.7 U	0.97 U	0.93 U	0.91 U	0.94 U
Phenanthrene, 3,6-dimethyl-	0.97 U	1.8 U	0.97 U	0.93 U	0.91 U	0.94 U
Fluoranthene	**9.0**	**29**	0.97 U	**7.0**	**26**	0.94 U
Pyrene	4.7 U	**17**	0.97 U	**6.1**	**12**	0.94 U
2-Methylfluoranthene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Retene	0.97 U	11 U	0.97 U	0.93 U	0.91 U	0.94 U
Benzo[a]anthracene	0.97 U	**4.2**	0.97 U	0.93 U	**3.1**	0.94 U
Chrysene	0.97 U	**8.7**	0.97 U	0.93 U	**6.2**	0.94 U
Chrysene, 5-methyl-	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Benzo[b]fluoranthene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Benzo[k]fluoranthene	0.97 U	5.4 U	0.97 U	0.93 U	0.91 U	0.94 U
Benzo[e]pyrene	0.97 U	2.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Benzo[a]pyrene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U

Appendix B. (continued)

Species	Oyster		Littleneck Clam			Butter Clam
Tissue	Soft Parts	Soft Parts	Soft Parts	Soft Parts	Soft Parts	Soft Parts
Sampling Site	Bayview St. Park	Swinomish Ch.	Person Road	March Point	Crandall Spit	Samish Island
Collection Date	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99	1-Jun-99
Sample Number	228030	228031	228032	228033	228034*	228035
PAH (ug/Kg)						
Perylene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Indeno[1,2,3-cd]pyrene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Dibenzo[a,h]anthracene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Benzo[ghi]perylene	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
C1-Naphthalenes	**6.8** NJ	**15** NJ	3.9 U	6.4 U	3.6 U	3.7 U
C2-Naphthalenes	3.9 U	**12** NJ	3.9 U	3.7 U	3.6 U	3.7 U
C3-Naphthalenes	3.9 U	**12** NJ	3.9 U	3.7 U	3.6 U	3.7 U
C4-Naphthalenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C1-Fluorenes	3.9 U	7.0 U	3.9 U	3.7 U	3.6 U	3.7 U
C2-Fluorenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C3-Fluorenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C1-Dibenzothiophenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C2-Dibenzothiophenes	3.9 U	2.5 U	3.9 U	3.7 U	3.6 U	3.7 U
C3-Dibenzothiophenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C1-Phenanthrenes/Anthracenes	3.9 U	17 U	3.9 U	3.7 U	3.6 U	3.7 U
C2-Phenanthrenes/Anthracenes	3.9 U	21 U	3.9 U	3.7 U	3.6 U	3.7 U
C3-Phenanthrenes/Anthracenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C4-Phenanthrenes/Anthracenes	3.9 U	11 U	3.9 U	3.7 U	3.6 U	3.7 U
C1-Fluoranthene/Pyrene	3.9 U	7.4 U	3.9 U	3.7 U	3.6 U	3.7 U
C1-Chrysenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C2-Chrysenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C3-Chrysenes	3.9 U	7.6 U	3.9 U	3.7 U	3.6 U	3.7 U
C4-Chrysenes	0.97 U	1.9 U	0.97 U	0.93 U	0.91 U	0.94 U
Butyltins (ug/Kg)						
Tetrabutyltin	1.8 U	1.7 U	1.8 U	1.8 U	2.1 U	2.2 U
Tributyltin chloride	**2.7**	**7.9**	1.8 U	1.8 U	2.1 U	2.2 U
Dibutyltin dichloride	1.8 U	1.7 U	1.8 U	1.8 U	2.1 U	2.2 U
Monobutyltin trichloride	1.8 U	2.1 U	1.8 U	2.5 U	2.1 U	2.2 U
Percent Lipids	0.9	1.0	0.3	0.3	0.3	0.4
Percent Solids	17	16	10	13	13	14

Appendix B. (continued)

Species	Dungeness Crab					
Tissue	Muscle	Muscle	Muscle	Muscle	Hepatopancreas	Hepatopancreas
Sampling Site	Samish Island	Hat Island	Fidalgo Bay	March Point	Samish Island	March Point
Collection Date	26-May-99	26-May-99	26-May-99	26-May-99	26-May-99	26-May-99
Sample Number	218020	218021	218022	218023	218024	218025
Metals (ug/Kg)						
Arsenic	**5700**	**8390**	**5230**	**7350**	na	na
Lead	**29** J	**33 J**	**11** J	**20** J	na	na
Cadmium	25 U	25 U	25 U	25 U	na	na
Selenium	**591**	**692**	**512**	**496**	na	na
Mercury	**70**	**75**	**41**	**56**	na	na
Pesticides (ug/Kg)						
Alpha-BHC	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Beta-BHC	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Gamma-BHC (Lindane)	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Delta-BHC	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Heptachlor	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Aldrin	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Heptachlor Epoxide	0.25 UJ	0.22 UJ	0.26 UJ	0.21 UJ	na	na
Trans-Chlordane (Gamma)	0.64 U	0.56 U	0.66 U	0.53 U	na	na
Endosulfan I	0.25 UJ	0.22 UJ	0.26 UJ	0.21 UJ	na	na
Dieldrin	0.25 U	0.22 U	0.26 U	0.21 U	na	na
4,4'-DDE	**0.25**	**0.19**	0.26 U	**0.22**	na	na
Endrin	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Endosulfan II	0.25 U	0.22 U	0.26 U	0.21 U	na	na
4,4'-DDD	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Endrin Aldehyde	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Endosulfan Sulfate	0.25 U	0.22 U	0.26 U	0.21 U	na	na
4,4'-DDT	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Endrin Ketone	0.25 UJ	0.22 UJ	0.26 UJ	0.21 UJ	na	na
Methoxychlor	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Alpha-Chlordene	0.32 UJ	0.28 UJ	0.33 UJ	0.27 UJ	na	na
Gamma-Chlordene	0.32 UJ	0.28 UJ	0.33 UJ	0.27 UJ	na	na
Oxychlordane	0.32 U	0.28 U	0.33 U	0.27 U	na	na
DDMU	0.32 U	0.28 U	0.33 U	0.27 U	na	na
Cis-Chlordane (Alpha)	0.25 U	0.22 U	0.26 U	0.21 U	na	na
Cis-Nonachlor	0.32 U	0.28 U	0.33 U	0.27 U	na	na
Kelthane (Dicofol)	1.3 U	1.1 U	1.3 U	1.1 U	na	na
2,4'-DDE	0.32 U	0.28 U	0.33 U	0.27 U	na	na
Trans-Nonachlor	0.32 U	0.28 U	0.33 U	0.27 U	na	na
2,4'-DDD	0.32 U	0.28 U	0.33 U	0.27 U	na	na
2,4'-DDT	0.32 U	0.28 U	0.33 U	0.27 U	na	na
Mirex	0.32 U	0.28 U	0.33 U	0.27 U	na	na
Toxaphene	13 U	11 U	13 U	11 U	na	na
4,4'-Dichlorobenzophenone	1.3 U	1.1 U	1.3 U	1.1 U	na	na
Hexachlorobenzene	0.32 U	0.28 U	0.33 U	0.27 U	na	na
Pentachloroanisole	0.32 U	0.28 U	0.33 U	0.27 U	na	na
Tetradifon (Tedion)	1.3 U	1.1 U	1.3 U	1.1 U	na	na
DCPA (dacthal)	0.64 U	0.56 U	0.66 U	0.53 U	na	na
Diazinon	64 U	56 U	66 U	53 U	na	na
Ethion	25 U	22 U	26 U	21 U	na	na
Parathion	1.3 U	1.1 U	1.3 U	1.1 U	na	na
Methyl Parathion	1.3 U	1.1 U	1.3 U	1.1 U	na	na
Treflan (Trifluralin)	1.3 U	1.1 U	1.3 U	1.1 U	na	na
Chlorpyriphos	0.64 U	0.56 U	0.66 U	0.53 U	na	na
Oxadiazon	1.3 U	1.1 U	1.3 U	1.1 U	na	na

Appendix B. (continued)

Species	Dungeness Crab					
Tissue	Muscle	Muscle	Muscle	Muscle	Hepatopancreas	Hepatopancreas
Sampling Site	Samish Island	Hat Island	Fidalgo Bay	March Point	Samish Island	March Point
Collection Date	26-May-99	26-May-99	26-May-99	26-May-99	26-May-99	26-May-99
Sample Number	218020	218021	218022	218023	218024	218025
PCBs (ug/Kg)						
PCB - 1260	2.5 U	2.2 U	2.6 U	2.1 U	na	na
PCB - 1254	2.5 U	2.2 U	2.6 U	2.1 U	na	na
PCB - 1221	2.5 U	2.2 U	2.6 U	2.1 U	na	na
PCB - 1232	2.5 U	2.2 U	2.6 U	2.1 U	na	na
PCB - 1248	**1.4** J	**1.2** J	2.6 U	2.1 U	na	na
PCB - 1016	2.5 U	2.2 U	2.6 U	2.1 U	na	na
PCB - 1242	2.5 U	2.2 U	2.6 U	2.1 U	na	na
Dioxins & Furans (ng/Kg)						
2378-TCDF	**0.51** J	**0.49** J	0.39 U	0.78 U	na	na
2378-TCDD	0.80 U	0.40 U	0.41 U	0.57 U	na	na
12378-PeCDF	0.79 U	0.32 U	0.69 U	0.84 U	na	na
23478-PeCDF	0.69 U	0.54 U	0.53 U	0.51 U	na	na
12378-PeCDD	0.89 U	1.20 U	0.58 U	1.10 U	na	na
123478-HxCDF	0.58 U	0.76 U	0.45 U	0.49 U	na	na
123678-HxCDF	1.20 U	0.46 U	0.68 U	1.50 U	na	na
234678-HxCDF	0.67 U	0.53 U	0.64 U	1.10 U	na	na
123789-HxCDF	1.20 U	0.66 U	1.50 U	1.20 U	na	na
123478-HxCDD	0.93 U	0.64 U	0.57 U	0.62 U	na	na
123678-HxCDD	1.60 U	0.65 U	0.62 U	1.60 U	na	na
123789-HxCDD	0.76 U	0.38 U	0.70 U	0.65 U	na	na
1234678-HpCDF	1.50 U	0.55 U	0.98 U	1.30 U	na	na
1234789-HpCDF	0.53 U	1.00 U	0.58 U	0.98 U	na	na
12234678-HpCDD	1.20 U	1.70 U	0.65 UJ	1.00 U	na	na
OCDF	1.80 U	1.40 U	0.86 U	1.20 U	na	na
OCDD	1.20 UJ	3.10 U	2.60 U	2.50 U	na	na
PAH (ug/Kg)						
Naphthalene	0.90 U	2.4 U	0.91 U	0.94 U	0.99 U	0.99 U
2-Methylnaphthalene	0.90 U	4.0 U	0.91 U	0.94 U	0.99 U	0.99 U
1-Methylnaphthalene	0.90 U	0.44 U	0.91 U	0.94 U	0.99 U	0.99 U
1,1'-Biphenyl	1.2 U	0.92 U	3.6 U	3.8 U	1.7 U	1.8 U
2-Chloronaphthalene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
2,6-Dimethylnaphthalene	3.6 U	7.4 U	3.6 U	3.8 U	**10**	**8.0**
Acenaphthylene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Acenaphthene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Dibenzofuran	0.90 U	6.4 U	0.91 U	0.94 U	0.99 U	0.99 U
1,6,7-Trimethylnaphthalene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Fluorene	0.90 U	0.92 U	0.91 U	0.94 U	0.99 U	0.99 U
9H-Fluorene, 1-methyl-	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Dibenzothiophene	0.90 U	2.6 U	0.91 U	0.94 U	0.99 U	0.99 U
Phenanthrene	12 U	18 U	0.91 U	0.94 U	12 U	15 U
Anthracene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
2-Methylphenanthrene	0.90 U	3.6 U	0.91 U	0.94 U	0.99 U	0.99 U
1-Methylphenanthrene	1.8 U	3.6 U	0.91 U	0.94 U	0.99 U	0.99 U
4,6-Dimethyldibenzothiophene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Phenanthrene, 3,6-dimethyl-	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Fluoranthene	0.90 U	4.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Pyrene	3.0 U	3.2 U	0.91 U	0.94 U	0.99 U	0.99 U
2-Methylfluoranthene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Retene	3.4 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Benzo[a]anthracene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Chrysene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Chrysene, 5-methyl-	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Benzo[b]fluoranthene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Benzo[k]fluoranthene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Benzo[e]pyrene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Benzo[a]pyrene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U

Appendix B. (continued)

Species	Dungeness Crab					
Tissue	Muscle	Muscle	Muscle	Muscle	Hepatopancreas	Hepatopancreas
Sampling Site	Samish Island	Hat Island	Fidalgo Bay	March Point	Samish Island	March Point
Collection Date	26-May-99	26-May-99	26-May-99	26-May-99	26-May-99	26-May-99
Sample Number	218020*	218021	218022	218023	218024	218025
PAH (ug/Kg)						
Perylene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Indeno[1,2,3-cd]pyrene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Dibenzo[a,h]anthracene	1.5 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Benzo[ghi]perylene	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
C1-Naphthalenes	**4.9** NJ	6.6 U	3.6 U	3.8 U	**6.9** NJ	**7.3** NJ
C2-Naphthalenes	3.6 U	7.4 U	3.6 U	3.8 U	**13** NJ	**18** NJ
C3-Naphthalenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C4-Naphthalenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C1-Fluorenes	6.4 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C2-Fluorenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C3-Fluorenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C1-Dibenzothiophenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C2-Dibenzothiophenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C3-Dibenzothiophenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C1-Phenanthrenes/Anthracenes	8.7 U	10.8 U	3.6 U	3.8 U	3.9 U	4.0 U
C2-Phenanthrenes/Anthracenes	6.8 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C3-Phenanthrenes/Anthracenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C4-Phenanthrenes/Anthracenes	3.4 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C1-Fluoranthene/Pyrene	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C1-Chrysenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C2-Chrysenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C3-Chrysenes	3.6 U	7.4 U	3.6 U	3.8 U	3.9 U	4.0 U
C4-Chrysenes	0.90 U	1.8 U	0.91 U	0.94 U	0.99 U	0.99 U
Butyltins (ug/Kg)						
Tetrabutyltin	2.6 U	1.4 U	1.9 U	1.9 U	na	na
Tributyltin chloride	2.6 U	1.4 U	1.9 U	1.9 U	na	na
Dibutyltin dichloride	2.6 U	1.4 U	1.9 U	1.9 U	na	na
Monobutyltin trichloride	2.6 U	1.4 U	1.9 U	1.9 U	na	na
Percent Lipids	0.2	0.1	0.1	0.1	13.3	4.9
Percent Solids	17	17	17	18	18	25

Appendix B. (continued)

Species	Mussel	
Tissue	Soft Parts	Soft Parts
Sampling Site	March Point	Samish Island
Collection Date	1-Jun-99	1-Jun-99
Sample Number	228036	228037
Metals (ug/Kg)		
Arsenic	na	na
Lead	na	na
Cadmium	na	na
Selenium	na	na
Mercury	na	na
Pesticides (ug/Kg)		
Alpha-BHC	na	na
Beta-BHC	na	na
Gamma-BHC (Lindane)	na	na
Delta-BHC	na	na
Heptachlor	na	na
Aldrin	na	na
Heptachlor Epoxide	na	na
Trans-Chlordane (Gamma)	na	na
Endosulfan I	na	na
Dieldrin	na	na
4,4'-DDE	na	na
Endrin	na	na
Endosulfan II	na	na
4,4'-DDD	na	na
Endrin Aldehyde	na	na
Endosulfan Sulfate	na	na
4,4'-DDT	na	na
Endrin Ketone	na	na
Methoxychlor	na	na
Alpha-Chlordene	na	na
Gamma-Chlordene	na	na
Oxychlordane	na	na
DDMU	na	na
Cis-Chlordane (Alpha)	na	na
Cis-Nonachlor	na	na
Kelthane (Dicofol)	na	na
2,4'-DDE	na	na
Trans-Nonachlor	na	na
2,4'-DDD	na	na
2,4'-DDT	na	na
Mirex	na	na
Toxaphene	na	na
4,4'-Dichlorobenzophenone	na	na
Hexachlorobenzene	na	na
Pentachloroanisole	na	na
Tetradifon (Tedion)	na	na
DCPA (dacthal)	na	na
Diazinon	na	na
Ethion	na	na
Parathion	na	na
Methyl Parathion	na	na
Treflan (Trifluralin)	na	na
Chlorpyriphos	na	na
Oxadiazon	na	na

Appendix B. (continued)

Species	Mussel	
Tissue	Soft Parts	Soft Parts
Sampling Site	March Point	Samish Island
Collection Date	1-Jun-99	1-Jun-99
Sample Number	228036	228037
PCBs (ug/Kg)		
PCB - 1260	na	na
PCB - 1254	na	na
PCB - 1221	na	na
PCB - 1232	na	na
PCB - 1248	na	na
PCB - 1016	na	na
PCB - 1242	na	na
Dioxins & Furans (ng/Kg)		
2378-TCDF	na	na
2378-TCDD	na	na
12378-PeCDF	na	na
23478-PeCDF	na	na
12378-PeCDD	na	na
123478-HxCDF	na	na
123678-HxCDF	na	na
234678-HxCDF	na	na
123789-HxCDF	na	na
123478-HxCDD	na	na
123678-HxCDD	na	na
123789-HxCDD	na	na
1234678-HpCDF	na	na
1234789-HpCDF	na	na
12234678-HpCDD	na	na
OCDF	na	na
OCDD	na	na
PAH (ug/Kg)		
Naphthalene	0.85 U	3.8 U
2-Methylnaphthalene	0.85 U	3.8 U
1-Methylnaphthalene	0.85 U	3.8 U
1,1'-Biphenyl	3.4 U	3.8 U
2-Chloronaphthalene	0.85 U	0.95 U
2,6-Dimethylnaphthalene	3.4 U	3.8 U
Acenaphthylene	0.85 U	0.95 U
Acenaphthene	0.85 U	0.95 U
Dibenzofuran	0.85 U	0.95 U
1,6,7-Trimethylnaphthalene	0.85 U	0.95 U
Fluorene	0.85 U	0.95 U
9H-Fluorene, 1-methyl-	0.85 U	0.95 U
Dibenzothiophene	0.85 U	0.95 U
Phenanthrene	**15**	6.8 U
Anthracene	0.85 U	0.95 U
2-Methylphenanthrene	**4.2**	0.95 U
1-Methylphenanthrene	**2.5**	0.95 U
4,6-Dimethyldibenzothiophene	0.85 U	0.95 U
Phenanthrene, 3,6-dimethyl-	0.85 U	0.95 U
Fluoranthene	**10**	0.95 U
Pyrene	**6.6**	0.95 U
2-Methylfluoranthene	0.85 U	0.95 U
Retene	6.1 U	3.2 U
Benzo[a]anthracene	0.85 U	0.95 U
Chrysene	**12**	0.95 U
Chrysene, 5-methyl-	0.85 U	0.95 U
Benzo[b]fluoranthene	0.85 U	0.95 U
Benzo[k]fluoranthene	0.85 U	0.95 U
Benzo[e]pyrene	**8.0**	0.95 U
Benzo[a]pyrene	0.85 U	0.95 U

Appendix B. (continued)

Species	Mussel	
Tissue	Soft Parts	Soft Parts
Sampling Site	March Point	Samish Island
Collection Date	1-Jun-99	1-Jun-99
Sample Number	228036	228037
PAH (ug/Kg)		
Perylene	0.85 U	0.95 U
Indeno[1,2,3-cd]pyrene	0.85 U	0.95 U
Dibenzo[a,h]anthracene	0.85 U	0.95 U
Benzo[ghi]perylene	0.85 U	0.95 U
C1-Naphthalenes	**5.6** NJ	4.8 U
C2-Naphthalenes	3.4 U	3.8 U
C3-Naphthalenes	**5.6** NJ	3.8 U
C4-Naphthalenes	3.4 U	3.8 U
C1-Fluorenes	3.4 U	9.0 U
C2-Fluorenes	3.4 U	3.8 U
C3-Fluorenes	3.4 U	3.8 U
C1-Dibenzothiophenes	3.4 U	3.8 U
C2-Dibenzothiophenes	3.4 U	3.8 U
C3-Dibenzothiophenes	3.4 U	3.8 U
C1-Phenanthrenes/Anthracenes	**15** NJ	6.6 U
C2-Phenanthrenes/Anthracenes	**2.8** NJ	3.8 U
C3-Phenanthrenes/Anthracenes	3.4 U	3.4 U
C4-Phenanthrenes/Anthracenes	18 U	3.8 U
C1-Fluoranthene/Pyrene	3.4 U	3.8 U
C1-Chrysenes	**32** NJ	3.8 U
C2-Chrysenes	3.4 U	3.8 U
C3-Chrysenes	3.4 U	3.8 U
C4-Chrysenes	0.85 U	0.95 U
Butyltins (ug/Kg)		
Tetrabutyltin	na	na
Tributyltin chloride	na	na
Dibutyltin dichloride	na	na
Monobutyltin trichloride	na	na
Percent Lipids	0.8	1.0
Percent Solids	11	14

Note: Detections highlighted in **BOLD**
na = not analyzed (PAH only)
U = not detected at or above reported value
J = estimated value
UJ = not detected at or above reported estimated value
NJ = evidence the analyte is present; numerical result is an estimate

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Appendix C

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March 8, 2000

TO: Art Johnson
Environmental Assessment Program
Department of Ecology

FROM: Glen Patrick
Public Health Advisor
Department of Health

SUBJECT: DETERMINATION OF FURTHER SHELLFISH SAMPLING NEEDS IN PADILLA BAY BASED ON HUMAN HEALTH

Thank you for contacting the Office of Environmental Health Assessments (OEHA) concerning chemistry data recently collected for shellfish from Padilla Bay as summarized in the draft report: *Results of a Screening Analysis for Metals and Organic Compounds in Shellfish from Padilla Bay and Vicinity*.[i] In providing this report, we were asked to evaluate if detected chemical concentrations from these limited number of shellfish samples indicate the presence of a potential human health concern based on tribal shellfish consumption, and thus warrant more intensive chemical and species-specific sampling and analysis. Due to the screening nature of the data provided, a detailed health assessment was not conducted. The brief discussion that follows includes a review of the shellfish chemistry data, and an overview of the evaluation method. Results of the screening evaluation are then discussed, followed by our conclusions regarding additional environmental sampling and analysis.

I hope this information is useful. Please contact me at 236-3177, should you have any questions regarding the following assessment or the conclusions expressed below.

Data Review

Non-depurated bivalve shellfish and crab samples collected from Padilla Bay were analyzed for 130 chemicals, which included metals, pesticides, polychlorinated biphenyls (PCBs), dioxins and furans, polycyclic aromatic hydrocarbons (PAHs), and butyltin compounds. Dungeness crab samples were a composite of five legal sized males; oyster composite samples consisted of 20 individuals each, while clam and mussel composite samples consisted of 50 individuals each. A total of 30 chemicals were detected at concentrations above their analytical detection limits (Table 1). Chemical detection varied by species and between locations. Additionally, analytical results for detected lead ,PCB, 2,3,7,8-TCDF, and some PAH concentrations were qualified as estimates.

Evaluation Method

To evaluate whether additional environmental sampling should be conducted in Padilla Bay, a screening evaluation based on human health toxicity endpoints was conducted in a manner consistent with the approach outlined by the United States Environmental Protection Agency (EPA).[ii] Screening chemical concentration values (SV) were calculated for 23 chemicals having existing human health toxicity criteria out of the 30 chemicals detected in shellfish from Padilla Bay (Table 2). Screening values, calculated by equations (1) and/or (2) depending on the availability of toxicity criteria, reflect a daily chemical intake level which is unlikely to result in any adverse human health impacts over ones lifetime of exposure. Toxicity criteria values developed by OEHA for mercury and DDT were used in place of EPA values.[iii,iv] Additionally, 10 percent of reported total arsenic concentrations were assumed to be of the toxic inorganic form.

Non-cancer endpoints: $SV_x = [(RfD_x \text{ x } BW) / (CR / UCF)] \text{ x } UCF$ (1)

Cancer endpoints: $SV_x = \{[(RL / SF_x) BW] / (CR / UCF)\} \text{ x } UCF$ (2)

Where:

- SV_x = Screening value for chemical x (ug/kg)
- RfD_x = Reference dose, oral for chemical x (mg/kg/day)
- BW = Body weight (kg)
- CR = Consumption rate for species of concern (g/day)
- RL = Risk Level (no units)
- SF_x = Slope factor, oral for chemical x (/mg/kg/day)
- UCF = Unit conversion factor (1000)

Screening calculation parameter values are listed in Table 3. To enhance the relevance of this evaluation to high shellfish consumers such as tribal members, species-specific shellfish consumption rates from the Tulalip Tribe, located in Marysville, Washington, were used. Determination of individual shellfish species consumption rates was performed using Stata™ and individual fish consumption survey respondent data provided by the Tulalip Tribe of Indians.[v] Ninetieth percentile shellfish consumption rates for Tulalip Tribal men were assumed for all chemicals with the exception of mercury. Ninetieth percentile shellfish consumption rates for Tulalip Tribal women of childbearing years (age 18 – 49) were used for mercury, since the most sensitive toxicological endpoint for mercury is impaired neurological development and long-term and/or delayed adverse health outcomes in children prenataly exposed. Additionally, consumption of fish and shellfish is the primary source of mercury exposure for most persons.[vi]

Tulalip Tribal member mean body weights of 86 kg and 75 kg were assumed for men and women respectively, along with a diminimus cancer risk level of $1 \text{x} 10^{-5}$. Since tribal shellfish consumption rates vary by species, five separate consumption rates were calculated for males and females who consume these particular species. Crab hepatopancreas data were not evaluated due to lack of consumption information.

Results

Out of the 130 chemicals sought in shellfish samples from Padilla Bay, 30 chemicals were detected. Chemical-specific toxicity criteria were available for 23 of the detected chemicals. Based on a comparison of detected chemical concentrations with calculated screening values for each of the species tested, two chemicals were identified as being of potential human health concern. Arsenic concentrations detected in all shellfish samples except mussels, which were not analyzed for arsenic, exceeded calculated screening values. The reported concentration of 2,3,7,8-TCDF (furan) in the Dugeness crab sample from Samish Island was also at the calculated screening value based on a dioxin toxicity equivalency (TEQ) factor of 0.1 for this compound. Chemicals not detected were assumed to not be of human health concern due to presumed low human exposure. Detected chemicals with toxicity criteria, other than arsenic and 2,3,7,8-TCDF, are also assumed to not be of human health concern. No conclusions are drawn for detected chemicals that lack existing toxicity criteria.

Conclusions

Arsenic concentrations reported for oysters, littleneck clams, and butter clams from this study are comparable to concentrations reported for bivalve shellfish from other areas in Puget Sound, including areas typically regarded as reference locations.[vii] However, arsenic concentrations reported for Dungeness crab from Padilla Bay are greater than the other shellfish species tested. Since arsenic is known to occur naturally in various geologic formations in and around Puget Sound at high concentrations, the issue of arsenic in Puget Sound shellfish is problematic and one of on-going research and discussion within this office. To help reduce uncertainties associated with assessing the human health implications of arsenic in shellfish, it is recommended that further sampling and analysis of shellfish for arsenic be conducted. Such sampling and analysis should include frequently consumed shellfish species and the measurement of both total arsenic and speciated arsenic during different seasons of the year using state-of-the-art analytic techniques, since shellfish arsenic toxicity is primarily associated with its inorganic species.

Reported furan concentrations were near the analytical detection limit for this compound and thus were qualified as estimates. Reported crab muscle tissue furan concentrations from this study are comparable to concentrations in crab reported by the Puget Sound Estuary Program for various urban embayments in Puget Sound and therefore may reflect the ambient concentration of this compound in Dungeness crab within Puget Sound.[viii] While scientists and health professionals worldwide acknowledge that exposure to dioxins and furans should be minimized, there is currently no agreement on what level of exposure constitutes a tolerable daily intake. Given the limitations of our toxicological understanding of this compound presently, and the background level of reported concentrations, additional sample collection and analysis for dioxins and furans is not recommended at this time in Padilla Bay.

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[i] Johnson, A. 2000. Results of a screening analysis for metals and organic compounds in shellfish from Padilla Bay and vicinity. Draft report. Washington State Department of Ecology, Olympia, Washington.

[ii] EPA. 1995. Guidance For Assessing Chemical Contaminant Data For Use in Fish Advisories: Volume 1, Fish Sampling and Analysis, Second Edition. Office of Water. EPA 823-R-95-007.

[iii] Department of Health. 1999. Evaluation of evidence related to the development of a tolerable daily intake for mercury. Office of Environmental Health Assessments, Olympia, Washington.

[iv] Department of Health. 1997. DDT and DDE transmission through breast milk: Yakima River basin. Office of Environmental Health Assessments, Olympia, Washington.

[v] Toy, K., Polissar, N., Liao, S., and Mittelstaedt, G. 1996. A fish consumption survey of the Tulalip and Squaxin Island tribes of the Puget Sound region. Tulalip Tribes, Department of Environment, 7615 Totem Beach Road, Marysville, WA 98271.

[vi] Bolger, M. 1995. Methylmercury (MeHg) – Hazard and risk. National Forum on Mercury in Fish, Proceedings. EPA 823-R-95-002.

[vii] Department of Health. 1996. Puget Sound Ambient Monitoring Program: 1992 and 1993 shellfish chemical contaminant data report. Washington State Department of Health, Olympia, Washington.

[viii] Puget Sound Estuary Program. 1991. Dioxin and furan concentrations in Puget Sound crabs. Prepared by PTI Environmental Services, Bellevue, Washington for the U.S. Environmental Protection Agency, Region 10, Office of Coastal Waters. EPA Contract 68-D8-0085.

Table 1. Chemicals Detected in Padilla Bay Shellfish Above Detection Limits*

Species		Oyster		Littleneck Clam			Butter Clam	Dungeness Crab (muscle)				Mussel	
Sample Site		Bayview St. Par	Swinomish Ch.	Person Rd.	March Point	Crandall Spit	Samish Is.	Samish Is.	Hat Is.	Fidalgo Bay	March Pt.	March Pt.	Samish Is.
Sample #	Units	228030	228031	228032	228033	228034	228035	218020	218021	218022	218023	228036	228037
Metals	*ug/kg*												
Arsenic		1460	1360	2520	2040	2310	2600	5700	8390	5230	7350		
Lead		55	43	51	128	104	46	29	33	11	20		
Cadmium		822	1460	211	317	410	366						
Selenium		660	470	750	535	674	599	591	692	512	496		
Mercury		21	22	21	15	26	11	70	75	41	56		
Pesticides	*ug/kg*												
Dieldrin		0.25	0.65										
4,4'-DDE		1.4						0.25	0.19		0.22		
4,4'-DDD		0.3	0.27										
4,4'-DDT		0.54	0.2										
Trans-Nonachlor		0.18	0.45										
PCBs	*ug/kg*												
1254			1.7										
1248		2.7	1.3		2.3			1.4	1.2				
Dioxins/Furans	*ug/kg*												
2378-TCDF		0.00069	0.00046					0.00051	0.00049				
PAHs	*ug/kg*												
2-Methylnaphthalene			11									4.2	
1-Methylnaphthalene			3.6									2.5	
1,1'-Biphenyl						1.8							
1,6,7-Trimethylnaphthalene			4.2										
Phenanthrene			19			24						15	
Fluoranthene		9	29		7	26						10	
Pyrene			17		6.1	12						6.6	
Benzo(a)anthracene			4.2										
Benzo(e)anthracene												8	
Chrysene			8.7			3.9						12	
C1-Naphthalenes		6.8	15					4.9				5.6	
C2-Naphthalenes			12										
C3-Naphthalenes			12									5.6	
C1-Phenanthrenes												15	
C2-Phenanthrenes												2.8	
C1-Chrysenes												32	
Butyltins	*ug/kg*												
Tributyltin Chloride		2.7	7.9										

* For data qualifiers see: Johnson, A. 2000. Results of a screening analysis for metals and organic compounds in shellfish from Padilla Bay and vicinity. Draft report. Washington State Department of Ecology, Olympia, Washington. Appendix C.

Table 2. Chemical and Species Specific Screening Values for Chemicals Detected in Padilla Bay Shellfish

Chemical	Reference Dose (RfD) (mg/kg/day)	Slope Factor (SF) (/mg/kg/day)	Oyster (ug/kg) Non-Cancer	Oyster (ug/kg) *Cancer*	Littleneck Clam (ug/kg) Non-Cancer	Littleneck Clam (ug/kg) *Cancer*	Butter clam (ug/kg) Non-Cancer	Butter clam (ug/kg) *Cancer*	D. Crab (muscle) (ug/kg) Non-Cancer	D. Crab (muscle) (ug/kg) *Cancer*	Mussel (ug/kg) Non-Cancer	Mussel (ug/kg) *Cancer*
Metals												
Arsenic @	3.00E-04	1.5	4623.18	*102.74*	4141.76	*92.04*	1134.72	*25.22*	442.81	*9.84*	8016.21	*178.14*
Lead	#											
Cadmium	1.00E-03		8110.85		7266.24		1990.74		776.86		14063.53	
Selenium	5.00E-03		40554.25		36331.22		9953.70		3884.28		70317.65	
Mercury (methyl)	8.00E-05 *	Females	272.73		937.50		212.01		121.95		2803.74	
Pesticides												
Dieldrin	5.00E-05	16	405.54	*5.07*	363.31	*4.54*	99.54	*1.24*	38.84	*0.49*	703.18	*8.79*
4,4'-DDE	5.00E-03 *	0.34	40554.25	*238.55*	36331.22	*213.71*	9953.70	*58.55*	3884.28	*22.85*	70317.65	*413.63*
4,4'-DDD		0.24		*337.95*		*302.76*		*82.95*		*32.37*		*585.98*
4,4'-DDT	5.00E-03 *	0.34	40554.25	*238.55*	36331.22	*213.71*	9953.70	*58.55*	3884.28	*22.85*	70317.65	*413.63*
Trans-Nonachlor	5.00E-04	0.35	4055.43	*231.74*	3633.12	*207.61*	995.37	*56.88*	388.43	*22.20*	7031.76	*401.82*
PCBs												
1254	2.00E-05	2	162.22	*40.55*	145.32	*36.33*	39.81	*9.95*	15.54	*3.88*	281.27	*70.32*
1248		2		*40.55*		*36.33*		*9.95*		*3.88*		*70.32*
Dioxins/Furans												
2378-TCDF ^^		150000 **		*0.00541*		*0.00484*		*0.00133*		*0.00052*		*0.00938*
PAHs												
2-Methylnaphthalene	2.00E-02		162217.02		145324.89		39814.81		15537.12		281270.58	
1-Methylnaphthalene	2.00E-02		162217.02		145324.89		39814.81		15537.12		281270.58	
1,1'-Biphenyl	5.00E-02		405542.54		363312.22		99537.04		38842.81		703176.46	
1,6,7-Trimethylnaphthalene	#											
Phenanthrene	#											
Fluoranthene	4.00E-02		324434.03		290649.78		79629.63		31074.25		562541.17	
Pyrene	3.00E-02		243325.52		217987.33		59722.22		23305.69		421905.88	
Benzo(a)anthracene		0.73 **		*111.11*		*99.54*		*27.27*		*10.64*		*192.65*
Benzo(e)anthracene	#											
Chrysene		0.0073 **		*11110.75*		*9953.76*		*2727.04*		*1064.19*		*19265.11*
C1-Naphthalenes	2.00E-02		162217.02		145324.89		39814.81		15537.12		281270.58	
C2-Naphthalenes	2.00E-02		162217.02		145324.89		39814.81		15537.12		281270.58	
C3-Naphthalenes	2.00E-02		162217.02		145324.89		39814.81		15537.12		281270.58	
C1-Phenanthrenes	#											
C2-Phenanthrenes	#											
C1-Chrysenes	#											
Butyltins												
Tributyltin Chloride	3.00E-04		2433.26		2179.87		597.22		233.06		4219.06	

@ RfD for inorganic arsenic, 90% of arsenic assumed to be of an organic species
* RfD value from Washington State Department of Health, Office of Environmental Health Assessments.
** SF from EPA HEAST tables (http://www.epa.gov/reg3hwmd/)
^^ Screening concentration for TCDF in terms of TCDD toxicity equivalents (TEQ) (TCDD x 10)
\# No RfD or SF available, chemical not assessed

Table 3. Parameter Values Used in Calculation of Padilla Bay Chemical Specific Screening Values (Consumption rates are 90th percentile values for Tulalip Tribal shellfish consumers)

Parameter		**Species**	**Males**	**Females (18-49)**
Body Wt. (kg)	BW		86.00	75.00
Risk Level (no units)	RL		1.00000E-05	1.00000E-05
Consumption Rate (g/d):	CR-o	Oyster	10.6	22.00
	CR-ln	LN Clam	11.8	6.40
	CR-bc	B. Clam	43.2	28.30
	CR-dc	D. Crab	110.7	49.20
	CR-m	Mussel	6.1	2.14
Slope Factor (/mg/kg/day)	SF	Chemical specific		
Reference Dose (mg/kg/day)	RfD	Chemical specific		