Contaminant Residues in Demersal Fish, Invertebrates, and Deployed Mussels in Selected Areas of the Puget Sound, WA

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Introduction

A clear understanding of contaminant levels in biota, the potential impact from exposure, and how contaminants accumulate and biomagnify through the food web is needed to protect threatened and endangered species, make meaningful decisions about cleanup of contaminated sediments and hazardous waste disposal sites, accurately assess and implement control of persistent, bioaccumulative, and toxic (PBT) pollutants, and assure the healthiness of seafood harvested from the Puget Sound. Ultimately, the effectiveness of cleanup and restoration programs, success of pollution abatement initiatives, and the potential impact of stormwater and other sources of contaminants on the health of the Puget Sound will be reflected in the well-being and condition of the plants and animals that live, feed, and forage in the waters of Puget Sound.

To obtain data on contaminant residues and the biological condition of demersal fish and macro-invertebrates from selected areas of the Puget Sound, The Puget Sound Ambient Monitoring Program (PSAMP) and the Puget Sound Naval Shipyard & Intermediate Maintenance Facility (PSNS&IMF) Project ENVironmental inVEStment (ENVVEST) partnered to increase the data yield from fish trawl surveys conducted by the Washington State Department of Fish and Wildlife (WDFW) in 2003 and 2005. PSAMP is a multi-agency effort to monitor the spatial and temporal trends in contaminant exposure in Puget Sound fish and macro-invertebrates (WDFW 2003). Project ENVVEST is a cooperative watershed project being conducted by PSNS&IMF, the Environmental Protection Agency, the Washington State Department of Ecology, and other participating technical stakeholders to develop Total Maximum Daily Loads (TMDL), assess ecological risks, and improve the environmental quality of Sinclair and Dyes Inlets and its surrounding watershed (Johnston 2004).

Methods

ENVVEST biota sampling consisted of obtaining additional specimens from the PSAMP trawls and conducting a caged mussel study in Sinclair and Dyes Inlets (Johnston et al. 2005). During the 2003 and 2005 PSAMP surveys, samples of English sole (*Parophrys vetulus*), rock sole (*Lepidopsetta bilineata*), sand sole (*Psettichthys melanostictus*), ratfish (*Hydrolagus colliei*), surfperch (*Cymatogaster aggregata*), staghorn sculpin (*Leptocottus armatus*), graceful crab (*Cancer gracilis*), and sea cucumber (*Parasticopus californicus*) were collected from monitoring stations in the Puget Sound (Figure 1). In Sinclair Inlet (Figure 2), caged mussels (*Mytilus galloprovincialis*) were deployed from June – September 2005 (84 days) at locations adjacent to the Shipyard and at reference locations within Sinclair and Dyes Inlets. Indigenous mussels (*Mytilus* sp.) were also collected from three stations in Sinclair Inlet (Applied Biomonitoring 2007). In addition, data on polychlorinated biphenyls (PCB) levels in fillets from English sole from 2003 were provided by WDFW, and data on residues of PCBs and metals in blue mussels (*Mytilus* spp.) sampled from Puget Sound Mussel Watch (MW) stations in 2000, 2002, and 2004 were obtained from the National Status and Trends Program (NOAA 2007).

The ENVVEST samples from the 2003 and 2005 PSAMP survey and 2005 caged mussel study were analyzed for residues of PCBs, metals, and lipid content. Tissues were homogenized as individual whole body specimens (except for shiner surfperch which were composites of 2-3 fish) including shell for crabs and excluding shells for mussels. Samples for metals analysis were freeze-dried, milled, and about 500 mg was digested with nitric

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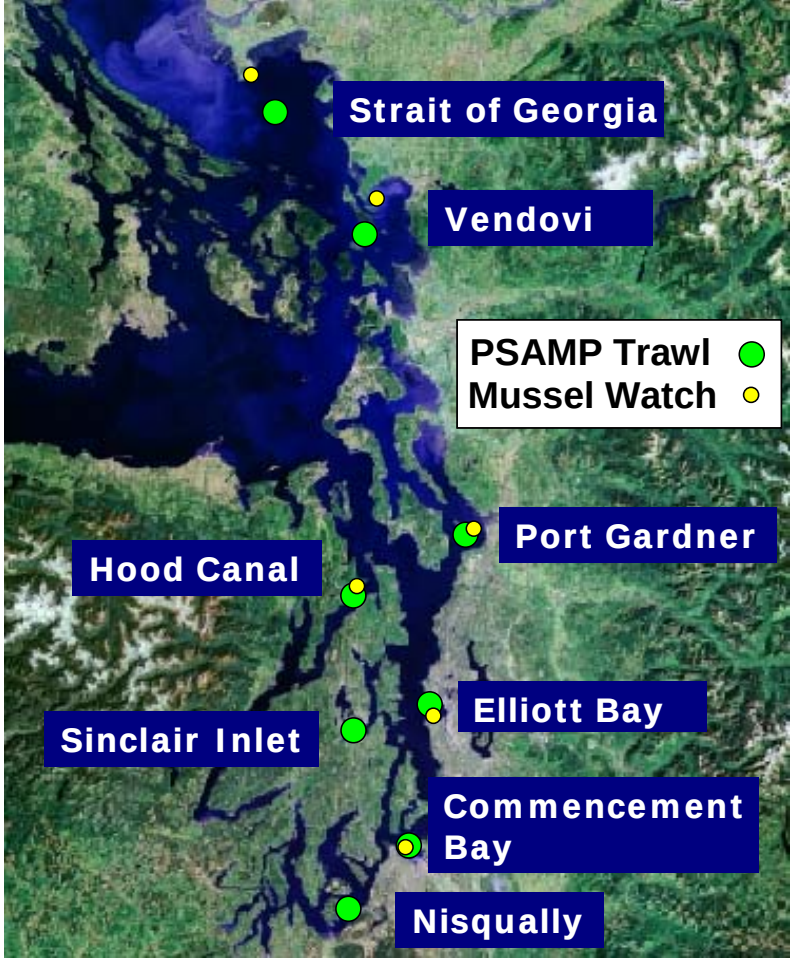

Figure 1. Location of PSAMP trawl and mussel watch stations in Puget Sound.

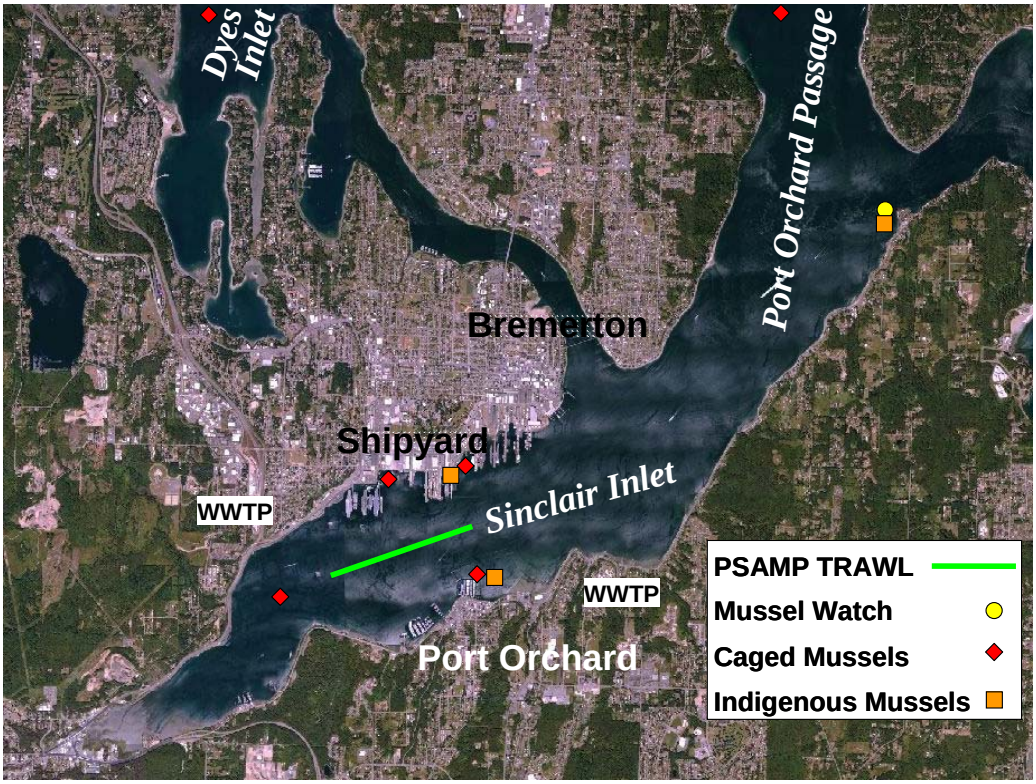

Figure 2. Locations of PSAMP trawl, mussel watch, caged mussel, and indigenous mussel stations in Sinclair and Dyes Inlets.

and hydrochloric acids for a minimum of 8 hr. Digested samples were analyzed for total Hg by CVAA; Ag, As, Cd, Cu, Ni, Pb, and Zn were analyzed using ICP-MS; and Cr was analyzed using ICP-OES. For PCB analysis about 15 – 30 g of wet tissue was spiked with surrogate compounds and extracted three times with methylene chloride. The combined extract was dried over anhydrous sodium sulfate, concentrated, and processed through an alumina cleanup procedure by GPC/HPLC. The 2003 ENVVEST samples were analyzed by GC/ECD to quantify 26 congeners (BMSL 2004). The 2005 ENVVEST samples were quantified using GC/MS methodology based on EPA Method 1668A to obtain data on 26 congeners as well as the 10 homologs (Brandenberger et al. 2006a; b). English sole samples from the 2003 PSAMP survey were processed by WDFW as skin-off fillets with aliquots from 20 individuals composted into each sample. These samples were analyzed for lipids and PCBs using an HPLC/PDA technique that quantified 15 congeners and estimated Total PCB (J. West, WDFW, personal communication). The mussel watch samples were analyzed for lipids, 18 PCB congeners by GC/ECD, and metals (NOAA 2007).

The sum of the ten homologs measured by GC/MS provided a definitive measure of Total PCB: Total PCB = ΣHCL_i, where HCL_i is the homolog concentration at each level of chlorination and i=1-10. A regression between Total PCB and the sum of the congeners measured in the same samples showed that Total PCB could be estimated: Total PCB = $2\times\Sigma PCB_i$, where PCB_i is the concentration of the measured congeners (Figure 3). The homolog distribution among the species analyzed varied (Figure 4). Hexachlorobiphenyls were the most abundant fraction, accounting for about 30-38% of the PCBs for all species; however, ratfish had higher fractions of tetrachlorobiphenyl (22%) and mussels had higher trichlorobiphenyl (22%).

An estimate of Total PCB in MW and WDFW samples was obtained using twice the sum of measured congeners. Although 26 congeners were quantified in the ENVVEST samples versus 18 for MW and 15 for WDFW, the additional ENVVEST congeners mainly consisted of very low concentrations of coplanar dioxin-like congeners that did not contribute very much to the sum. Additionally, all groups contained the most abundant congeners and the estimate of Total PCBs from congener data is consistent with other studies (O'Conner 2002). The PCB data from the English sole fillets (PCB_{Fillet} ng/g wet) were converted into whole body concentrations (PCB_{WB} ng/g wet weight) by assuming that the PCB concentration of the whole body was proportional to the lipid content of the fillet (f_lipid_{Fillet} g lipid/g wet tissue) : $PCB_{WB} = PCB_{Fillet}\times 3.44$, where 3.44 was the ratio of the average whole body lipid content measured in English sole (f_lipid_{WB}, average = 0.0193, n=51) to average fillet lipid (average = 0.0056, n=49). Similarly, PCBs in skin-off fillets were estimated from the whole body concentration: $PCB_{Fillet} = PCB_{WB}/3.44$.

Tissue Residue Benchmarks

Tissue residue benchmarks were developed to assess the potential for ecological and human health effects (Table 1). Ecological benchmarks consisted of water quality criteria (WQC) -based tissue screening values (TSV) and bioaccumulation critical values (B_{CV}), critical body residues corresponding to the no observed effect dose (NOED) and the lowest observed effect dose (LOED) for a fish or invertebrate species, and dietary benchmarks were set to the No Observed Adverse Effect Level (NOAEL) for the consumption of prey by black ducks, ospreys, and harbor seal pups. Benchmarks for seafood consumption of PCBs and Hg were protective of recreational and Tribal fishers (WDOH 2006).

The TSV and B_{CV} benchmarks based on WQC were developed by back-calculating the tissue residue that would occur if the water exposure was set to the chronic value, assuming bioconcentration factors (BCF) available from the literature were applicable (Table 1A): TSV or $B_{CV} = WQC_{Chronic} \times BCF$. Originally developed for conducting screening level ecorisk assessments at Navy sites (Shepard 1998; MESO-E 2000; URS 2002), the TSVs were calculated using the lowest freshwater or saltwater criteria in effect when the TSVs were developed (1996) and lipid-based BCFs that were assumed to be applicable to all aquatic species. Subsequently, TSV values for Ag, Cu, and Zn were recalculated to account for metal bioavailability (Dyer et al. 2000). The B_{CV} benchmarks were calculated using the most recent saltwater WQC (USEPA 2002) and BCFs specific to marine invertebrates and fishes. The WQC benchmarks were generally lower than the tissue residue

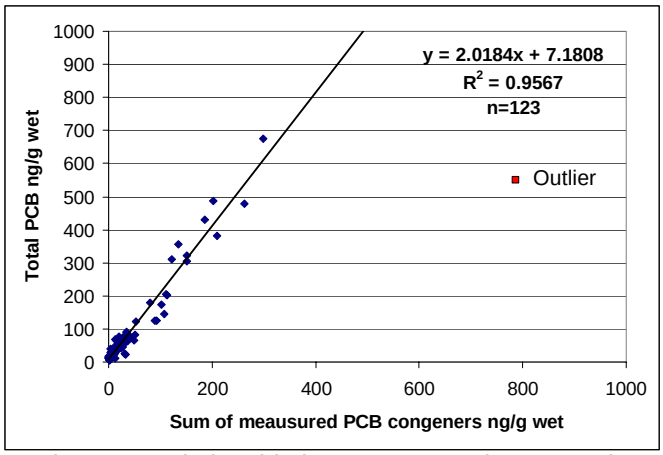

Figure 3. Relationship between sum of measured congeners and Total PCB by sum of homologs.

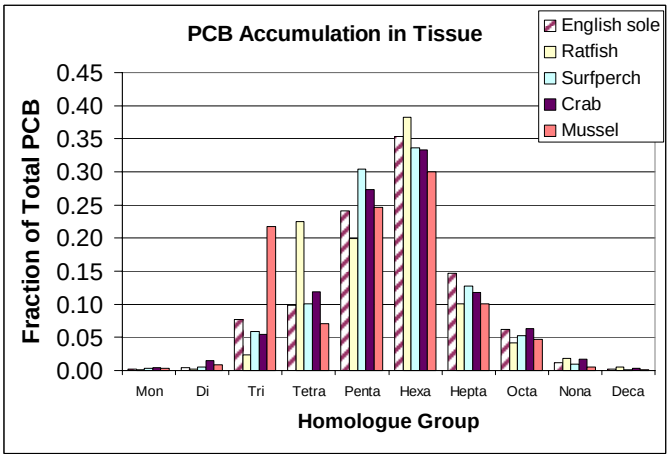

Figure 4. Distribution of PCB homologs measured in fish and invertebrates.

Table 3. Benchmarks (ug/g wet weight) of ecological (A) and human health (B) effects for residues in fish and invertebrate tissues, or both (All). Blank indicates data not available to support benchmark development; * indicates UF applied.

A. Ecological Tissue Residue Benchmarks

	Water Quality Based			Critical Body Residues				Dietary Exposure[5]		
	TSV[1]	B_{CV}[2]		NOED[3]		LOED[4]		D_{BDuck}[6]	D_{Osprey}[7]	$D_{SealPup}$[8]
	All	Fish	Invert.	Fish	Invert.	Fish	Invert.	All	Fish	All
Chemical	ug/g	ug/g	ug/g	ug/g	ug/g	ug/g	ug/g	ug/g	ug/g	ug/g
Ag	0.37							18.44 u*	9.22 u*	
As	1.58							57.11 v	28.56 v	
Cd	0.04	0.60	37.20	0.40 a	0.90 b	0.90 c	1.29 d	16.11 v	8.06 v	
Cr	0.18	0.80	20.00	0.18 e	0.14 f*	0.44 g*	1.44 h*	11.11 w	5.56 w	
Cu	3.00	0.62	12.40	1.68 i	3.40 j	1.96 i	4.00 k	522.22 x	261.11 x	68.25 aa
Hg	0.12	4.69	19.65					5.00 y	2.50 y	5.83 aa
Ni	0.39	0.39	16.40		2.83 l*		28.30 m*	860.00 v	430.00 v	
Pb	0.06	0.40	81.00	2.55 n	4.00 o	4.02 n	20.35 p*	12.56 y	6.28 y	
Zn	20.00	3.81	1620.00					161.11 x	80.56 x	
Total PCB	0.44	3.02	0.94	1.50 q	0.60 r	2.20 s	1.10 t	2.00 z	1.00 z	0.80 aa

B. Human Health Benchmarks

	NonCancer		Cancer	
	Rec[9]	Tribe90[10]	Rec[9]	Tribe90[10]
Chemical	ug/g	ug/g	ug/g	ug/g
Hg	0.64 bb	0.50 bb		
Total PCB	0.13 cc	0.10 cc	0.74 dd	0.25 dd

a Spehar et al. 1978, flagfish survival

b Rule and Alden 1996, grass shrimp survival

c Meteyer et al. 1988, sheepshead minnow dev.

d Carr et al. 1985, mysid growth

e Buhler et al. 1977, rainbow trout physiology

f Poulton et al. 1989, stonefly ED10×(UF=0.1)

1 Tissue Screening Value (TSV) based on water quality criteria (WQC) and bioaccumulation factors for aquatic species (URS 2002, Dyer et al. 2000).

2 Bioaccumulation Critical Values (Bcv) based on current chronic saltwater WQC and BCFs for marine fish (USEPA 2005) and bivalves (Thoman et al 1999).

3 NOED is the highest tissue residue that did not cause an effect to marine and freshwater fish and invertebrates. Footnoted references cited in ERED 2004.

4 LOED is the lowest tissue residue that caused an effect to marine and freshwater fish and invertebrates. Footnoted references cited in ERED 2004.

5 Concentraton in prey equal to NOAEL assuming 100% of diet obtained from single prey. Footnoted references cited in Sample et al. 1996.

6 Black duck (D_{BDuck}) NOAEL assuming body weight (bw) = 1.25 kg, food consumption rate (*f*) = 125 g/day, lifespan (L) = 1.0

7 Osprey (D_{Osprey}) NOAEL assuming bw = 1.5 kg, food consumption rate *f* = 300 g/day, L = 1.0

8 Harbor seal pup ($D_{SealPup}$) NOAEL assuming bw = 20.6 kg (Ross et al. 2004), *f* = 2.24 kg/day (2×adult *f*/bw), L = 1.0

9 Based on average ingestion rates for English Sole (11 g/day) by recreational 70 kg adult fishers with EF=365 day and ED=30 yr (WDOH 2006).

10 Based on 90th percentile ingestion rates for English Sole (14.07 g/day) by Suquamish Tribe 90 kg adult fishers with EF=365 day and ED=70 yr (WDOH 2006).

g Buhler et al. 1977, rainbow trout phys. ED50×(UF=0.5)

h Poulton et al. 1989, stonefly ED10×(UF=0.75)

i Hansen et al. 2002, rainbow trout fry dev.

j St.-Jean et al. 2003, juvenile mussel growth

k Grout and Levings 2001, mussel growth

l Wilson 1983, clam survival ED50×(UF=0.05)

m Wilson 1983, clam survival ED50×(UF=0.5)

n Holcombe et al. 1976, brook trout dev.

o Sundelin 1984, amphipod survival

p Ritterhoff and Zuake 1997, copepod surv. LC50×(UF=0.5)

q Hansen et al. 1975, sheepshd. minnow surv.

o Velduizen- and Holwerda 1991, mussel surv.

s Hansen et al. 1974, pinfish survival

t Hansen et al. 1974, grass shrimp survival

u Van Vleet 1982, mallard duckling 4 wk NOAEL×(UF=0.2)

v Sample et al. 1996, mallard duck NOAEL

w Sample et al. 1996, black duck NOAEL

x Sample et al. 1996, chicken NOAEL

y Sample et al. 1996, japensese quail NOAEL

z Sample et al. 1996, ringneck pheasant NOAEL

aa Sample et al. 1996, mink NOAEL

bb Hg Reference Dose (RfD) = 0.0001 mg/kg/day (WDOH 2006)

cc PCB RfD=0.00002 mg/kg/day WDOH '06

dd PCB cancer slope factor CSF= 2 $(mg/kg\text{-}day)^{-1}$

concentration that would be expected to cause an effect; therefore, residue levels below the TSV are assumed to pose little or no risk to aquatic biota (Shepard 1995, URS 2002, Dyer et al. 2000).

Critical body residues are defined as the threshold concentration of a contaminant in the tissue of an organism above which adverse effects could occur (McCarty et al. 1992, Pabst 1999). Generally, the effect occurs as a result of narcosis (non-cancer effects) and can result in mortality (acute effects) or a reduction in fecundity, reproduction, or growth (chronic effects). Data from the US Army Corps of Engineers Environmental Residue-Effects Database (ERED 2002) were used to develop benchmarks for critical body residues. The database was searched for effects on reproduction, growth, development, and survival. Results that were based on adult or juvenile exposure, whole body concentration, and ingestion or absorption were used, if available. Benchmarks were selected for the LOED and highest NOED (without exceeding LOED) for fish and invertebrates (Table 1). Uncertainty factors (UF) were used to make other effect levels (EL_{ERED}, e.g., ED_{50}, ED_{25}, etc.) comparable to NOEDs and LOEDs (U.S. EPA 1995, Sample et el. 1996): NOED = $EL_{ERED} \times UF$; LOED = $EL_{ERED} \times UF$.

Dietary benchmarks for avian and mammal predators were derived from toxicity values of similar species for black ducks (*Anas rubripes*), ospreys (*Pandion haliaetus*), and harbor seal pups (*Phoca vitulina*), the most sensitive life stage for seals (Table 1A). The NOAELs for mink (*Mustela vison* - $NOAEL_{mink}$) were converted to effects levels for harbor seal pups ($NOAEL_{SealPup}$) by scaling the dose to the ratio of mink body weight (bw) to the bw of harbor seal pups: $NOAEL_{SeaPup} = NOAEL_{Mink}(bw_{Mink}/bw_{SealPup})^{¼}$ (Sample et al. 1996). Due to the similarity in toxicity values reported for avian species (Sample et al. 1996), the NOAELs for black ducks and ospreys were assumed to be equivalent to the NOAELs reported for other avian species. The dietary benchmark (T_D) was calculated as the chemical dose in the food of a predator that would equal the NOAEL: $T_D = (NOAEL \times UF)/(aRfDL)$, where a = chemical assimilation factor (0.9 for all chemicals), R = food ingestion rate (g food/g bw day^{-1}), f = food consumption rate (g/day), D = fraction of diet (set to 1.0 for each prey item), and L = fraction of predator's life span (set to 1.0 for each predator) (Sample et al 1996).

Seafood benchmarks were developed with the exposure parameters used to determine the health risks of average recreational adult fishers (Rec) and the ninetieth percentile of Suquamish Tribal adult fishers (Tribe90) from consumption of Puget Sound seafood (Table 1B, WDOH 2006). The benchmarks were obtained by back calculating the concentration of PCBs and Hg in edible tissues (S_C) equal to the allowable risk levels established by WDOH (2006) for non-cancer: $S_C = RfD(BW \times AT_{NC})/(IR \times CF \times EF \times ED)$ and cancer endpoints: $S_C = 10^{-4}/CSF \times (BW \times AT_{NC})/(IR \times CF \times EF \times ED)$, where *RfD* is the non-cancer reference dose (mg/kg/day, 0.00002 for Total PCB and 0.0001 for Hg), BW is adult body weight (70 kg), AT_{NC} is the non-cancer averaging time (days, 10950 for Rec and 25550 for Tribe90), IR is the ingestion rate (g/day, 11 for Rec and 14.07 for Tribe90, based on English sole ingestion rates by Urban, Near Urban, and Non Urban populations), CF is the conversion factor 0.001, EF is the exposure frequency (365 d/yr), ED is the adult exposure duration (yr, 30 for Rec and 70 for Tribe90), 10^{-4} is the cancer risk threshold, *CSF* is the cancer slope factor (2 $(mg/kg\text{-}day)^{-1}$), and AT_C is the cancer averaging time (25550 days). Total PCB in edible tissues of fish were estimated using the whole body to fillet conversion factor calculated for English sole ($^1/_{3.44}$). Total PCB in edible crab meat was estimated from crab whole body concentration by assuming that the crab meat had the same lipid content as lobster claws and tail (0.47%, MESO-E 2000). Whole body concentrations of mussels and sea cucumbers were used as the edible portion for PCBs. Because Hg does not necessarily partition to lipids like PCBs, there is no readily available method for estimating concentrations of Hg in edible tissues from whole body concentrations. Therefore, the whole body Hg concentration for all species was used to provide a relative comparison.

Trophic Transfer

Whether contaminants are biomagnifing in the food web can be evaluated by calculating a Food Web Magnification Factor (*FWMF*) defined as $FWMF = e^b$, where b is obtained from a natural log-linear regression between contaminant concentration (C) and trophic level (TL) of species sampled from the food web:

Ln(C) = a + b(TL). A $FWMF > 1$ suggests biomagnification and $FWMF < 1$ suggests trophic dilution (Fisk et al. 2001, Mackintosh et al. 2004). TLs were assigned to the species sampled based on literature values (Fishbase 2007, UBC 2007) and regressions were calculated by sampling area (Figure 1).

Results and Discussion

The whole body (wet weight) concentrations of As, Ag, and Hg were highest in ratfish; crabs had the highest levels of Cu and Ni; mussels had the highest levels of Cd and Zn; sea cucumbers had the highest concentrations of Cr and Pb; and the highest levels of PCBs were found in English sole and ratfish (see Supplemental Information). Ratfish had much higher lipid levels on a whole body wet-weight basis, averaging 11% (range 7 – 18%) compared to an average of 3% (1 – 4%) for surf perch, 2% (0.5 – 6.7%) for English sole, 1.4% (0.7 – 2%) for mussels, about 1% for the other fishes and less than 1% for crabs and sea cucumbers. The urban areas had higher concentrations of contaminants than the other areas, particularly PCBs, Hg, Pb, and Zn. However, higher levels of Hg and As were measured in samples from the Strait of Georgia, and As was elevated in samples from Nisqually. High Cu levels in crab tissue is not unexpected because crabs and other malacostracan crustaceans have Cu-based blood, hemocyanin, and therefore have naturally high Cu levels (Barnes 1980).

Comparison to the ecological benchmarks for Total PCB showed that English sole from Sinclair Inlet, Elliot Bay, and Commencement Bay and ratfish from Sinclair Inlet exceeded the TSV, SealPup, and Osprey benchmarks. One ratfish sample from Sinclair Inlet also exceeded the NOED (Figure 5A). The PCBs measured in blue mussels showed an increasing gradient in concentration from the stations in the north of the Sound to the highest levels in Central and South Sound. The PCBs levels in English sole from Sinclair Inlet, Elliot Bay, and Commencement Bay were similar, but there were large differences in PCB concentrations measured in sea cucumber, crabs, ratfish, rock sole, surf perch, and sculpin collected from Sinclair Inlet compared to the reference locations. The PCBs in edible tissues of English sole from Sinclair Inlet, Elliot Bay, and Commencement Bay and ratfish and crabs from Sinclair Inlet exceeded seafood benchmarks for non-cancer exposure to recreational and tribal fishers (Figure 5B). These results are consistent with consumption warnings for English sole and other fish from the Puget Sound (WDOH 2006). The elevated PCB levels estimated for crab meats from Sinclair Inlet may be due to the relatively low lipid content of the crab samples (<0.4%) which could result in overestimating the amount of PCBs partitioned into the edible crab meat tissue.

The whole body concentrations of Hg were the highest in ratfish (Figure 5C), with maximum concentrations observed in specimens from the Strait of Georgia and Sinclair Inlet. Elevated Hg levels above the TSV were measured in samples of ratfish, rock sole, sand sole, sculpin, and mussels from Sinclair Inlet, English sole from Elliot Bay, Port Gardner, and Nisqually, and ratfish from the Strait of Georgia, Hood Canal, and Nisqually. Two of the three rock sole samples from Nisqually also exceeded the TSV. None of the mussel tissue samples exceeded the benchmarks, but the authors note that the Mussel Watch 2004 data (MW2004) may be biased high. Only two samples were at the Tribe90 benchmark of 0.5 ug/g: a ratfish sample from Strait of Georgia (0.52 ug/g) and English sole sample from Elliot Bay (0.45 ug/g). There is uncertainty about comparing Hg concentrations to the seafood benchmarks because it is unclear whether the whole body concentration over- or under- estimates the concentration in edible (fillet) tissues.

The ecological and seafood benchmarks are useful in evaluating the potential impact of individual chemicals, but equally important is the ability to assess the cumulative impacts of multiple chemicals as well as other environmental stressors. Results obtained from the caged mussel study help to address this issue because the impact of all environmental stressors on growth and survival, biomarkers of sublethal stress such as DNA damage, as well as contaminant residues can be evaluated. The results from the caged mussel study showed that PCBs, PAHs, and Pb were accumulated at elevated levels in Sinclair Inlet; however, the mussels remained healthy throughout the deployment and contaminants did not appear to accumulate to harmful levels (Applied Biomonitoring 2007; Steinert 2006). This approach in conjunction with monitoring levels of DNA damage which rapidly respond to environmental stressors could be used to further resolve temporal and spatial trends and identify the stressors responsible.

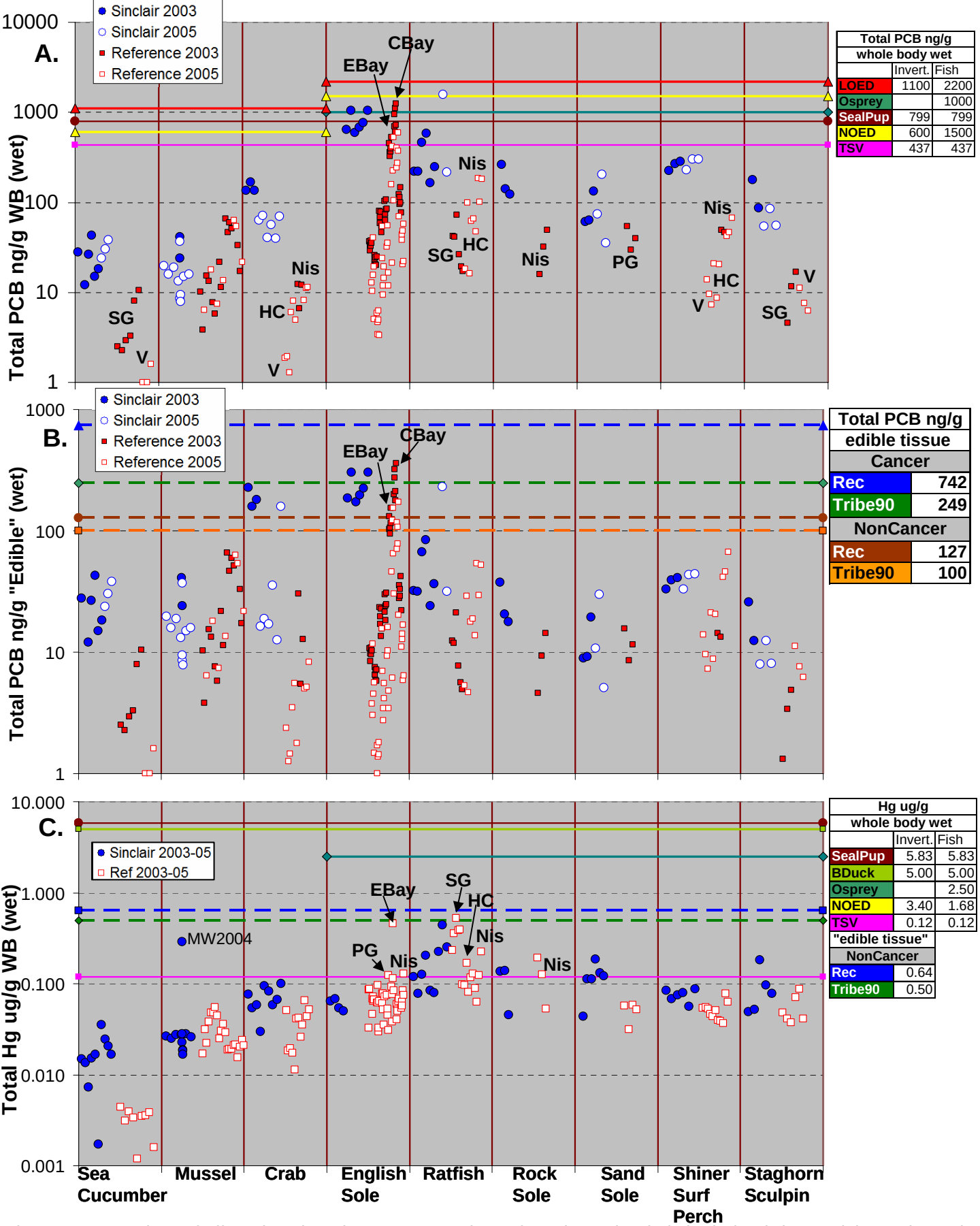

Figure 5. Comparison of effects benchmarks to concentrations of Total PCB in whole body (WB) tissues (A), Total PCB in edible tissues (B), and Hg in WB tissues (C) in species from Sinclair Inlet and reference areas. Data from reference areas for each species are plotted from left to right in order of north to south stations: Strait of Georgia (SG), Vendovi (V), Hood Canal (HC), Port Gardner (PG), Elliot Bay (EBay), Comm. Bay (CBay), and Nisqually (Nis).

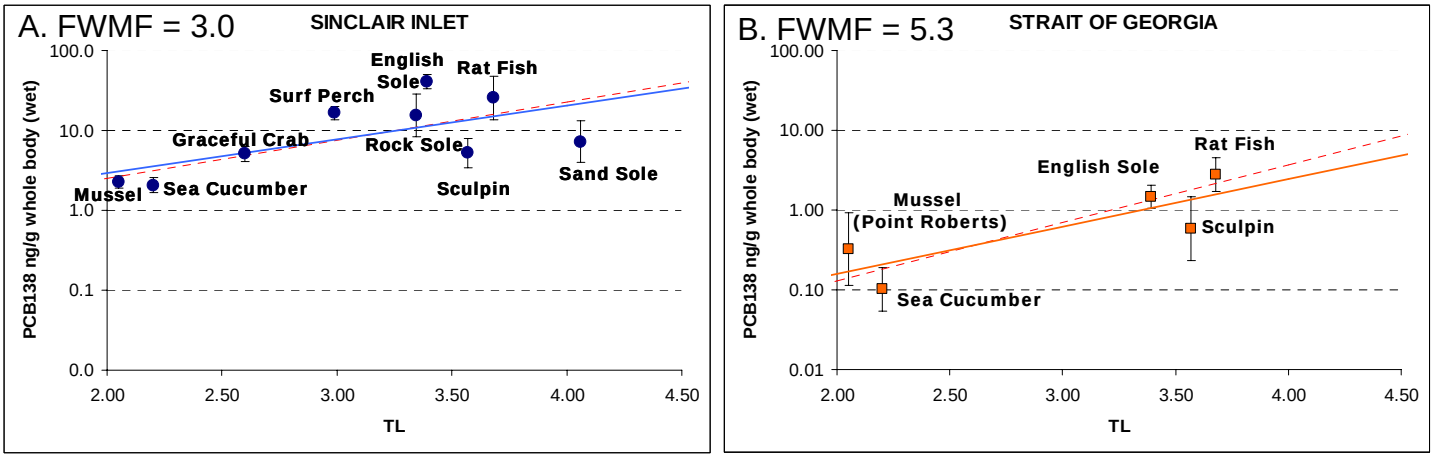

Figure 6. Regression between Trophic Level (TL) and mean concentration of PCB138 (solid line) for species sampled in Sinclair Inlet (A) and Strait of Georgia. Dashed line is regression using individual specimens.

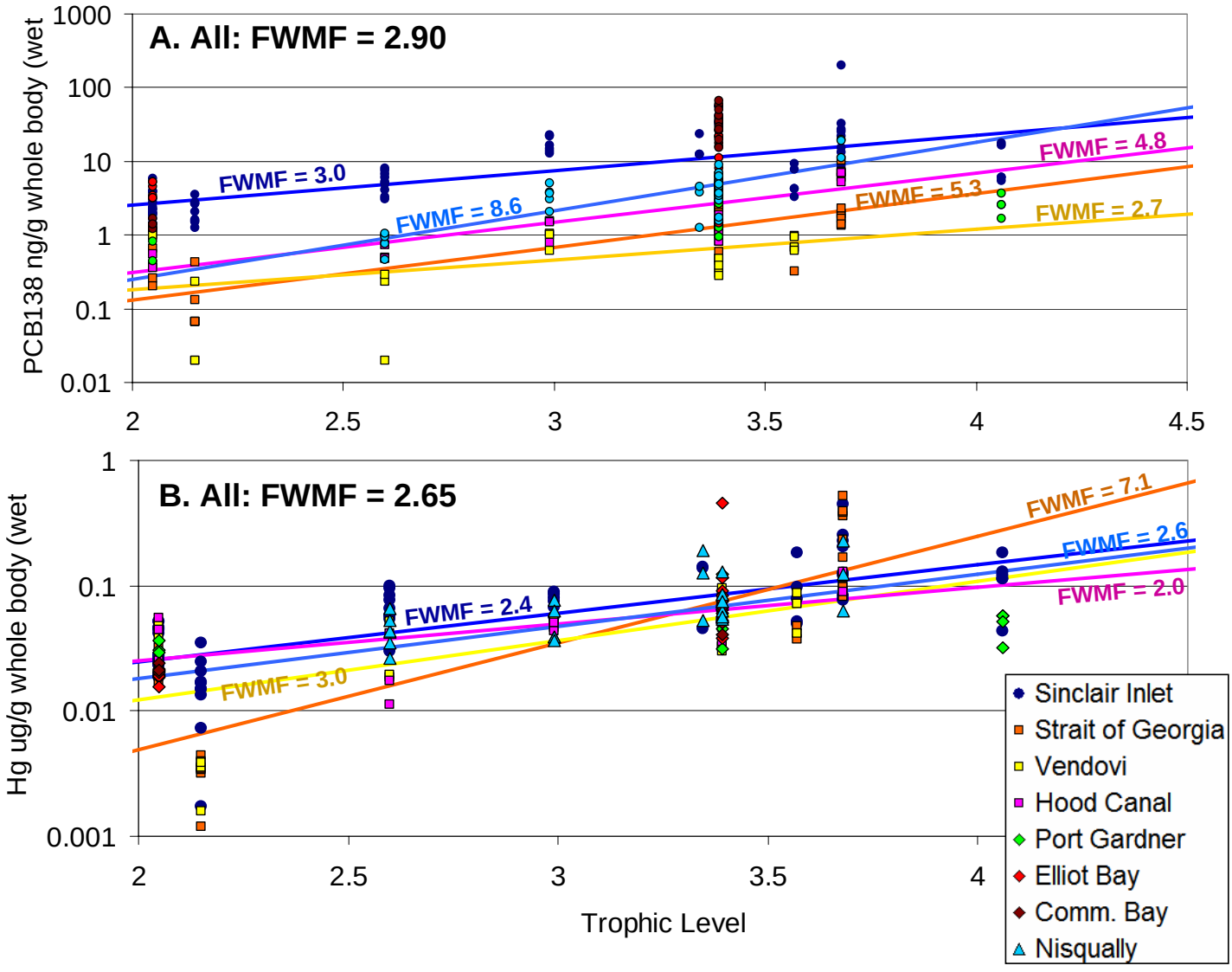

Figure 7. Relationship between Trophic Level and concentrations of PCB138 (A) and Hg (B) measured in species sampled from all of the Puget Sound. Lines show the regression results and Food Web Magnification Factors (FWMF) for specific areas of Puget Sound.

The trophic transfer evaluation resulted in FWMFs > 1 for As, Hg, some PCB congeners, and Total PCB suggesting that these chemicals are biomagnifing in areas of the Puget Sound. There was a relatively high variation in FWMF among areas of the Sound. For example, the FWMF for PCB138 from the Strait of Georgia was about a factor of 2 higher than the FWMF obtained for Sinclair Inlet (Figure 6) and ranged from 2.7 for Vendovi to 8.6 for Nisqually (Figure 7A). Similarly, FWMFs calculated for Hg varied from 2.0 to 7.1 (Figure 7B). This suggests that in certain regions of the Sound the food web is more efficient in transferring chemicals to higher trophic levels, signifying higher risk in those areas to top level predators such as marine mammals and sea birds. The differences in FWMFs may be due to differences in the sources, biogeochemical availability, and linkages in the food web that are operating differently in the various regions of the Sound. While these results are very intriguing, there is uncertainty in the findings because the number of samples from the food web are relatively small and the assumed TLs, based on literature values, may be quite different from the actual trophic position which may vary between and within regions of the Sound. Even slight changes in the TL of the species sampled could make a significant difference in the estimate of trophic transfer through the food web.

More data, especially information on ecological linkages within the Puget Sound food web, is needed to understand the trophic relationships and biogeochemical cycling of contaminants in the Sound. Critical to this effort is to obtain data on stable isotope ratios of C, N, and S for representative biota from regions of the Puget Sound. In other studies, stable isotope analysis has been conducted to estimate the trophic position of samples based on trophic fractionation of C and N (Fry 2006) and to assess if fishes and invertebrates feed primarily on pelagic or benthic prey by S ratios (Yamanaka et al. 2003). For example, studies in the Northern Gulf of Mexico (Snyder et al. 2007) have determined that trophic level can be inferred from the apparent enrichment of $\delta^{13}C$ and $\delta^{15}N$ relative to phytoplankton values. Additionally, depleted $\delta^{34}S$ relative to phytoplankton can be used to infer benthic rather than pelagic production because sulfate reduction in sediments results in lower $\delta^{34}S$ levels.

Conclusions

Contaminant levels in representative fish and invertebrates from various regions of the Puget Sound were evaluated to characterize tissue residue levels, assess potential ecological and human health impacts, and determine whether chemicals are being biomagnified in the food web. Clearly an ongoing monitoring framework is needed to provide a context for interpreting the spatial and temporal variations and identify possible sources and mechanisms of exposure to chemicals. The tissue residue benchmarks provide a means of evaluating exposure levels, and data on contaminant levels across the food web are needed to evaluate bioaccumulation. The data indicates that gradients of chemical exposure are present across the Puget Sound Region, potentially harmful exposures of some chemicals are present in certain areas, and some contaminants are biomagnifying in the food web.

Supplemental Information

Supplemental information can be downloaded from http://www.psmem.org/assets/PRIVATE/suppl_info.html.

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