

Polychlorinated Biphenyls in Cottids, Mussels, and Sediment in Southern Puget Sound, Washington

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Polychlorinated biphenyls (PCB) are a group of toxic, industrially prepared chlorinated hydrocarbons first identified in environmental samples by JENSEN (1966). Since then, the global distribution of PCB has been well documented (RISEBROUGH et al. 1968 and HAMMOND et al. 1972). We examined the intertidal distribution of PCB in Southern Puget Sound, and report herein the observed concentration of PCB in the mussel, Mytilus edulis; several species of cottids (bottom fish) including Leptocottus armatus and Oligocottus maculosus; and surface sediments. Puget Sound is a highly productive, complex network of channels and inlets with about 2,190 miles of shoreline located in the northwest corner of Washington State, U.S.A.

METHODS AND MATERIALS

Eighteen sites were chosen throughout Southern Puget Sound (Fig. 1), such that roughly one-half were located in harbor areas, close to population centers. The remainder of the sites were spaced evenly about Southern Puget Sound. Samples were collected between June and September 1975 during low tides. Mussels were taken from a relocatable study plot at each site. Cottids were seined immediately offshore and a portion of the top 2 cm of sediment was taken at the low tide mark. Biological samples were wrapped in baked aluminum foil and frozen until analysis.

Mollusks were shucked prior to analysis. A sufficient number of whole organism(s) to yield a minimum of 10 g of tissue (wet weight) were accurately weighed and digested in a minimum of 20 mls of B.F.M. solution (2 vols 70% perchloric acid in 3 vols glacial acetic acid) (STANLEY and LE FAVOURE 1965) for 4-6 hours over a steam bath. After cooling, the volume was doubled with distilled water. The sample was transferred to a separatory funnel and extracted with 3 vols of 20 ml each of glass-distilled hexane. Approximately 10 ml of the hexane extract was transferred to a centrifuge tube and shaken for 3 minutes with 5-10 grains of shiny copper turnings (previously soxhlet-extracted in hexane) to remove sulfur compounds. The copper turnings were removed and 1-2 ml of concentrated sulfuric acid was added to the hexane extract for cleanup (MURPHY 1972). The centrifuge tube was shaken for 1 minute and then centrifuged at 3000 rpm for 10 minutes. The sulfuric

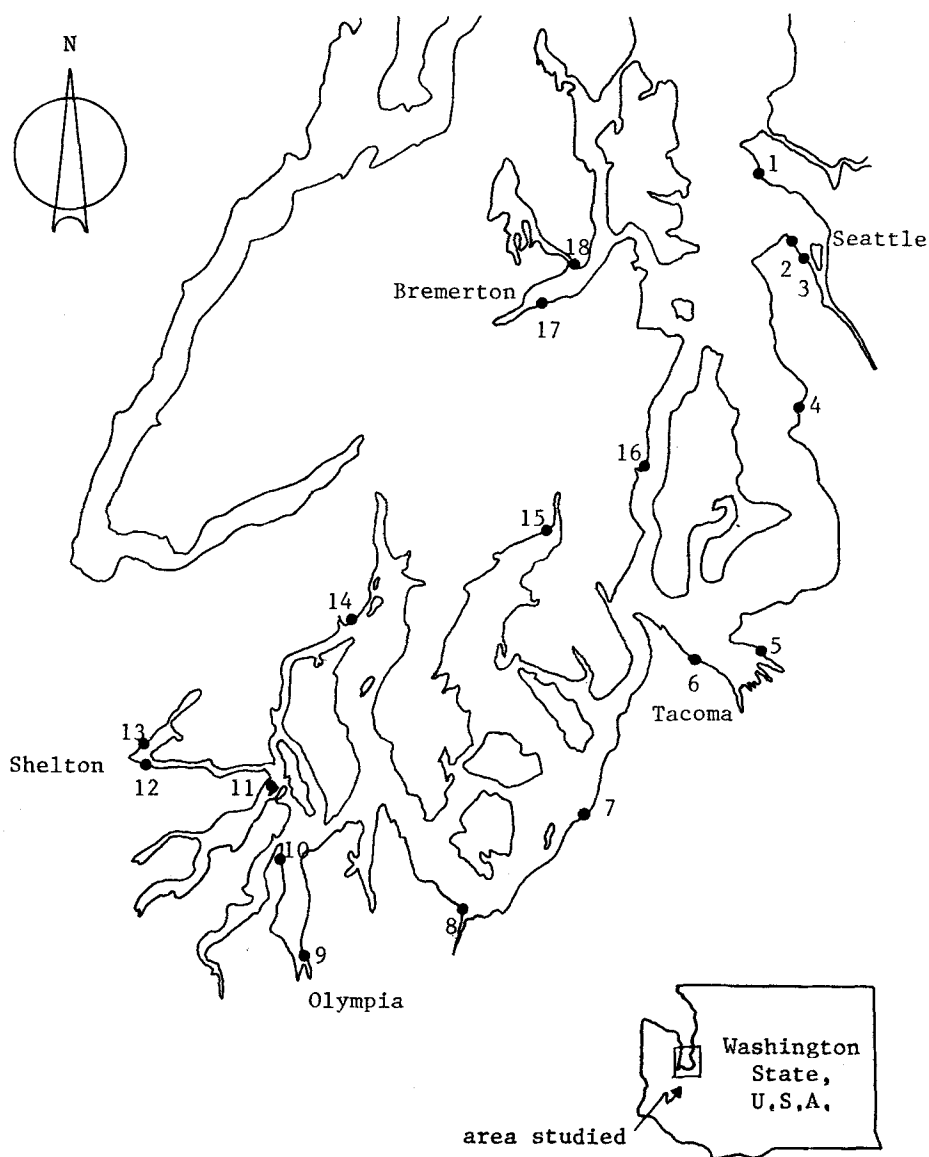


Figure 1- Site locations in Southern Puget Sound, Washington

acid portion was frozen with dry ice, and the hexane layer was removed and concentrated to a volume appropriate for injection.

Sediment samples were freeze-dried for approximately 48 hours. Approximately 60-100 g of freeze-dried sediment was transferred to a fritted-glass extraction thimble and soxhlet-extracted with approximately 120 ml glass-distilled hexane for 12-18 hours. Sulfur was removed from the hexane extracts either by passing the hexane through a sulfuric acid-celite column (DAVIDOW 1950) or by adding soxhlet-extracted copper turnings. The hexane extract was concentrated, cleaned up with concentrated sulfuric acid, and centrifuged as described above.

A Hewlett-Packard 5700A electron-capture gas chromatograph (^{63}Ni detector) coupled to a H.P. 3380A integrator was used to quantitatively determine the amount of PCB present in each sample. An appropriate volume of sample was injected into a coiled 6' glass column packed with 1" 33% NaOH/KOH on Gas Chrom Q 80/100 mesh (MILLER and WELLS 1969) followed by 10% DC-200 on Gas Chrom Q 80/100 mesh maintained at the following conditions: oven temperature, 225C; detector temperature, 300C; carrier gas, 60 ml/min 95% argon-methane.

Samples were quantified by individual homolog analysis. The mean weight percent of each homolog in a PCB standard (equal quantities of Monsanto Aroclor 1242, 1254, and 1260) was determined from figures published by WEBB and MCCALL (1973). Peaks were identified by comparing peak profiles and relative retention times in the PCB standard to those in the environmental samples. The levels of each homolog in environmental samples was calculated by quantitative comparison to standards injected daily. To compare levels among the different sites, 11 selected peaks were quantified in each sample. Figure 2 shows chromatograms of the PCB standard and two environmental samples with the 11 peaks used in quantification marked by arrows. The levels reported are the sum of the PCB in these 11 peaks representing about 70-80% of the total PCB present.

RESULTS

The concentration of PCB in cottid, mussel, and sediment samples taken from each of the 18 sites in Southern Puget Sound are given in Table 1.* For comparative purposes the levels reported at each site were adjusted to give equal importance to each type sample and summed. This figure, which we call the composite index, C.I., and the equation used to derive it are also given in Table 1.

The six sites showing the highest levels were located in harbor areas of the three largest urban centers in Southern Puget Sound. Three of the sites were located in Seattle (pop. 515,000), two in Tacoma (pop. 155,500), and one in Bremerton (pop. 36,000). Two of the three Seattle sites showed levels significantly higher than all the other sites. Site no. 2 at Armenie Beach displayed levels almost twice as high on our

* DDE [1,1-Dichloro-2,2-bis(p-chlorophenyl)ethylene] and HCB (hexachlorobenzene) were detected in our samples but were not quantified.

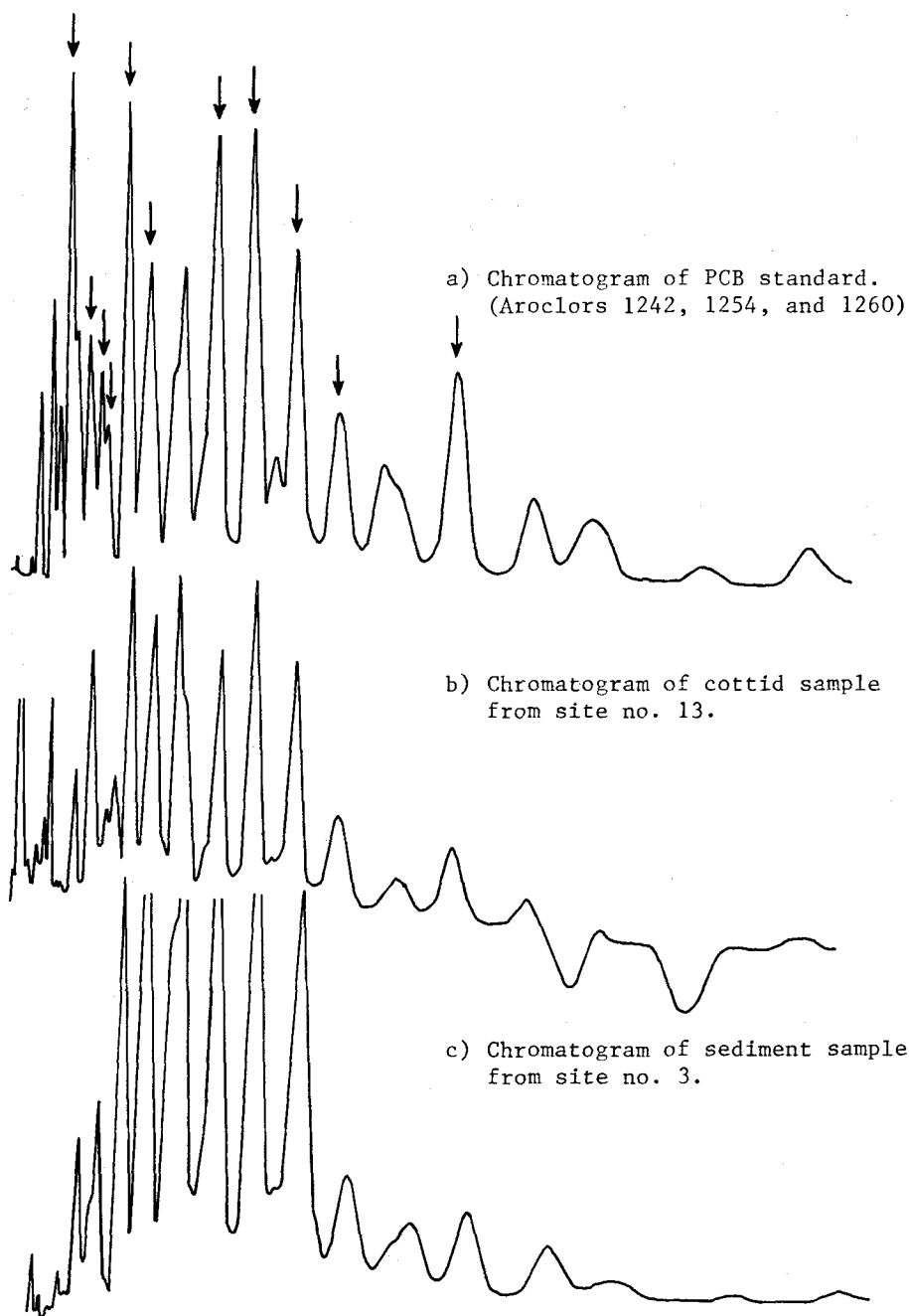


Figure 2- Chromatograms of PCB standard and cottid and sediment sample taken from Puget Sound with peaks that were quantified and summed to get total PCB levels marked by arrows.

TABLE 1

Concentration of PCB in Cottids, Mussels, and Sediment from Southern Puget Sound

Site no.	Concentration in cottids (ng/g wet wt)	Concentration in mussels (ng/g wet wt)	Concentration in sediment (ng/g dry wt)	C.I.*	Rank
1	65	85	1.8	467	5
2	470	95	70	1330	2
3	840	210	330	3945	1
4	180	31	1.0	307	8
5	200	72	6.3	531	4
6	500	38	17	769	3
7	100	16	1.5	174	11
8	130	50	2.6	348	7
9	56	27	1.1	172	12
10	66	30	2.1	204	10
11	29	14	0.7	90	17
12	29	24	1.5	135	13
13	62	11	3.1	129	14
14	21	16	0.7	90	18
15	29	10	5.7	108	16
16	63	11	1.8	119	15
17	160	14	10	223	9
18	190	40	7.3	400	6

* Composite Index = $C_i + (M_i)(\bar{C}/\bar{M}) + (S_i)(\bar{C}/\bar{S})$

C_i = cottid level M_i = mussel level S_i = sediment level

\bar{C} = mean of cottid levels \bar{M} = mean of mussel levels

\bar{S} = mean of sediment levels

composite index as any other site except site no. 3. Site no. 3 on the Duwamish River, an industrial area about 1 mile downstream from a point where over 200 gallons of a PCB-containing fluid had been spilled in September 1974 (DE YONGE 1974), was over five times as high as the other sites. The six sites showing the lowest PCB levels (bottom third) were located in areas of lowest population density. The six sites showing midrange levels of PCB (middle third) were dominated by moderately populated and industrialized areas but included one fairly isolated site near the mouth of the Nisqually River (site no. 8).

CONCLUSIONS

In Southern Puget Sound, PCB levels tend to be highest at or near the areas of highest population density, industry, and shipping activity. This is to be expected, since municipal and industrial outfalls provide the major input of PCB into the environment. PCB tends to accumulate near the source due to its low solubility in water and high affinity for sediment and particulate matter (HAMMOND et al. 1972). The levels of PCB that we found in our samples are comparable to those found in similar organisms from other coastal and estuarine systems throughout the world. Average PCB concentrations in mussels and herring from sites along the coast of Sweden were 30 and 270 ng/g (wet wt.), respectively (JENSEN et al. 1969). In the Irish coastal system, mussels were reported to have from 50 to 500 ng/g (wet wt.) and herring contained from 10 to 2000 ng/g (wet wt.) of PCB (HOLDGATE 1970). Mussels from the New Brunswick coast and herring from the Bay of Fundy contained 140 and 540 ng/g (wet wt.), respectively (ZITKO 1971). PCB levels in mussels from San Francisco Bay were reported to be between 30 and 60 ng/g (wet wt.) (RISEBROUGH and SCHMIDT 1975).

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