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The Distribution of Polychlorinated Biphenyls in the Sediments and Cottids of the Puyallup River System

A Spring Quarter Research Project

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INTRODUCTION

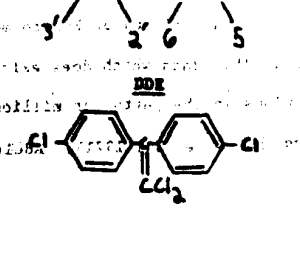
Polychlorinated biphenyls are a group of toxic aromatic compounds related to DDT presently found throughout terrestrial and marine environments. PCB's were first recognized as an environmental contaminant when Jensen identified them as a group of compounds interfering with pesticide analysis.(Jensen, 1966). Since Jensen's findings,several studies have documented the presence of PCBs in food chains throughout the global ecosystem. Risebrough and coworkers found PCBs to be widely distributed in fish and birds of Western North America, as well as in breading birds and their eggs from Antartica, Chile, Panama and other Southern Hemisphere regions (Risebrough et al., 1968). One year later Jensen and coworkers documented the presence of PCB in mussels, cod and dogfish of the Swedish coast; salmon, herring and grey seals of the Baltic sea; as well as in several marine organisms from the Stockholm archipelago, Gulf of Bothnia and the Gulf of Finland.(Jensen et al. 1969). PCBs have been found in the sediments and fish of all major river systems studied in the U.S. (Henderson, 1971). In Puget Sound PCBs have been found in both sediments and fish, with the highest levels occurring near areas of high industialization (Mowrer et al. 1977). Calambokidis and coworkers found high levels of PCB's in harbor seals in both Puget Sound and the outer Washington coast (Calambokidis et al., from the text of the presentation given January, 1978).

The lethal and sub-lethal effects of PCB to most organisims is still not clearly understood. That data which does exist suggests PCBs to be toxic to many organisms in the parts per million(ppm) and even parts per billion(ppb) range (Hansen et al. 1971). Adult female rhesus monkeys when

fed a diet of 5 ppm for 1.5 years were unable to reproduce, and those fed 2.5 ppm of PCB for the same time period, gave birth to under-weight infants who, soon after birth, developed facial acne, swelled eyelids, loss of facial hair and hyperpigmentation of the skin (Allen 1976). Mink as well have proven to be extremely sensitive to PCBs, encountering reproductive difficulties when fed a dietary level of 1 ppm PCB (Platonow 1973). The toxicity of PCB to fish has been shown to vary greatly between different species. Mebsker et al. found that young flagfish and fathead minnows encountered mortality and reduced growth when PCB levels in water approached 2.2 ppb (Mebsker et al. 1974). The channel catfish's 96-hour LC50 value for Arpelor 1248 (the amount of PCB necessary to produce 50% mortality after a 96-hr exposure time) has proven to be relatively high at 6,000 ppb, while that of the blue gill is much lower at 278 ppm (Stalling et al. 1972).

The molecular nature of PCB is such that close to 210 different compounds are possible due to the different possible arrangements of chlorine on the benzene ring of the PCB molecule (Cook 1972). A diagram of a PCB molecule is shown below. (DDE, which was also quantified in this study is shown for comparison). PCBs are sold commercially by the Monsanto Corp.

PCB



DDE

under the trade name Aroclor. There are five mixtures available, varying in chlorine content from 21% to 60% chlorine.

Such industrially desirable characteristics as nonflammability, chemical stability, high dielectric constant and plasticizing ability have made PCBs a highly regarded and used compound in industry. PCBs have been used in capacitors, transformers, hydraulic fluids, plasticizers and as a pesticide extender. In the past PCBs have also been used in the manufacture of sealants, adhesives, paints and printing inks. In 1977 the toxic substances control act was passed which provides for the eventual phasing out; over a two year period, of PCBs for all uses (Toxic Substances Control Act of 1977).

PCBs can enter the environment through a number of pathways. Unlike DDT, which was most often applied over large areas of land, PCB contamination of the envrironment usually generatas from a single point source. One such point source has proven to be municipal sewage discharges into rivers, lakes and coastal waters. Young and coworkers calculated that in 1975, 720 Kg of PCB was discharged into pacific coastal waters by five southern California sewage treatment plants (Young et el. 1975). Other known sources of PCB discharge into the environment are leaks from transformers, capacitors and hydraulic systems, spills in the manufacture and transportation of PCB ,vaporization into the atmosphere and disposal of PCB containing material into land fills (Nisbet and Sarofim 1972).

Although no reliable estimates are kept of the total amount of PCB released into the environment, one study done in 1970 by Nisbet and Sarofim calculated that between 1930 and 1970,30 thousand tons of PCBs were lost to the atmosphere, 60 thousand tons to fresh and marine waters and 300 thousand tons to dumps and land fills (Nisbet and Sarofim 1972).

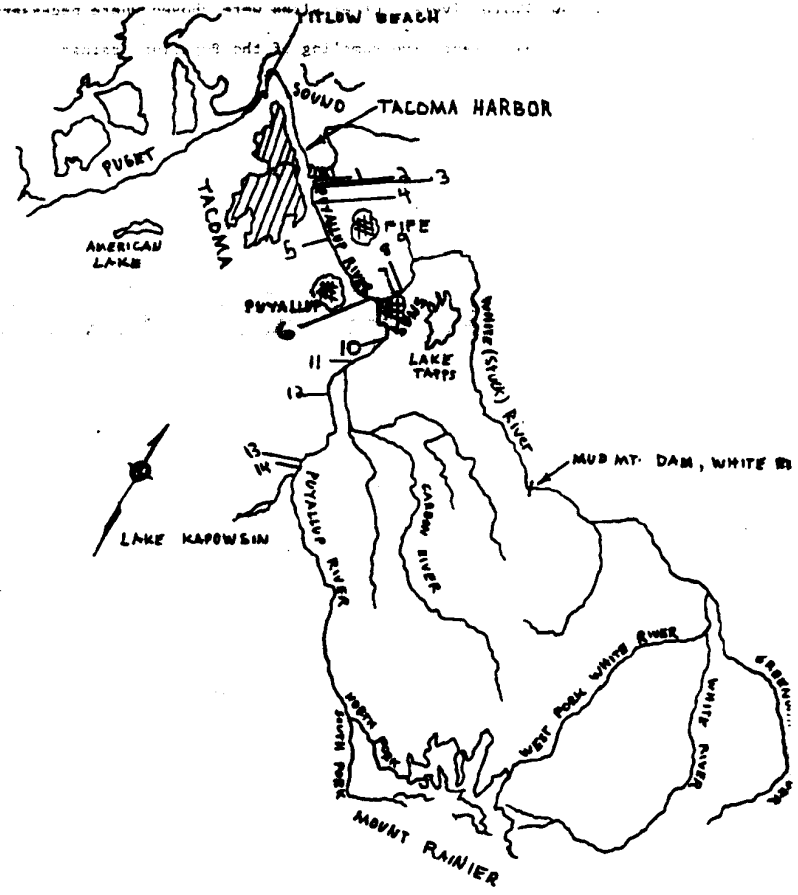
Once released into the environment there are two major transport modes PCB can follow. PCB released into the atmosphere is absorbed onto suspended particulates and then transported by the prevailing winds, eventually being deposited as fallout on terrestrial and marine environments. Through this kind of transport Nisbet and Sarofim estimate that 1,000 to 2,000 tons of PCB a year are deposited on to the terrastrial environment. A second possible route that PCB can follow in the environment is travel by water. Because of it's high affinity for sediments and low water solubility, most PCB discharged into the aquatic environment is deposited on bottom sediments and sludges with transport occurring by means of water born particulates (Nisbet and Sarofim 1972). These waterborn particulates have shown to be an effective medium by which fish can concentrate large quantities of PCB in their tissues. (Zitko 1974).

In 1975 a study was completed by students of The Evergreen State College which documented the distribution of PCB in the sediments and benthic organisms of Southern Puget Sound (Mowrer et al. 1977). The purpose of our study was to determine what impact, if any, the Puyallup river has on PCB levels in Commencement Bay, which is part of Puget Sound. To acheive this and we studied the distribution of PCB in the sediments and cottids at 14 sites on the Puyallup and White (Stuck) rivers (Figure 1). In addition, two western grebes were analyzed to provide data on biomagnification of PCBs in the food chain. At sites in Tacoma, Puyallup and Sumner, samples were taken to determine what contribution the municipal sewage treatment plants of these towns were making to the PCB load of the Puyallup river. Sites 1,7 and 8 were chosen to assess the PCB contribution of industry to the White and Puyallup rivers. Samples taken at site 9 on the White river and at site 13 on the Puyallup were done to assess the con-

tribution that power generating facilities make to the PCB load of the Puyallup and White rivers. Other sites were chosen where necessary to provide representative sampling of the Puyallup drainage.

Figure 1

SITE LOCATIONS



FIELD PROCEDURES

Materials and Equipment

The materials required for sampling each site included a 24" x 8" round minnow trap; 4' x 20' beach saina; cantigrade field thermometer; 4 oz. collection jar; metal spatula; aluminum foil; 100' cloth measuring tape; stop watch and fish

Methods

Sediments were collected by scooping 2 oz. of the top 3 cm of sediment directly into a clean collection jar from an average depth of 0.7 meters. Sediments were also collected by the use of a 6" x 6" x 12" grab sampler in an average depth of 1 m of water. The central portion of the sample was used for analysis to avoid any contamination from the sampler sides. Multiple sediment samples from each site were pooled for analysis. Samples were kept on ice until returned to the laboratory.

The collection of cottid samples was performed by the use of 24" x 8" minnow traps. The traps were baited, weighted, and placed on the river bottom in an average depth of 1 meter of water, 1 meter from shore. The traps were checked after 48 hours and in the absence of specimens, were rebaited and reset. A 40' x 20' beach seine was also used for collection, but with little success. Specimens obtained were wrapped in aluminum foil and placed on ice until returned to the laboratory.

LABORATORY PROCEDURES

Materials and Equipment

Pesticide quality glass-distilled hexane, acetone and 2,2,4-trimethylpentane were supplied by Burdick and Jackson laboratories. Concentrated sulfuric acid and copper turnings were supplied by J.T. Baker Chemical Company. G.F. Smith Chemical Company supplied BFM solution, Mixture F (2 parts 70% perchloric acid in 3 parts glacial acetic acid). Monsanto Chemical Co. supplied the polychlorinated biphenyls Aroclor 1242, 1254 and 1260.

Samples were analyzed on a Hewlett-Packard 5700A electron capture gas chromatograph (^{63}Ni detector) on line with a Hewlett-Packard 7123A chart recorder and with a Hewlett-Packard 3350A integrator or Perkin-Elmer Pep I Data System. Other equipment included a Hamilton Co. Inc. # 701 10 ul syringe; Mettler # 72 balance; Ohaus Dial-O-Gram balance; Büchi Rotovapor rotary evaporator; VWR - MSE GT-2 centrifuge; Thermovac Industries Freeze dryer; Thermodyne Type 2000 Furnace, Lindberge hot plate; Precision - Scientific vari heat; Buchler Instruments Thermo Lift; Corning PC-35 Hot Plate.

Methods

(1) Biological Specimens- The weight and length of each specimen was recorded. Samples were then placed into 125 ml beakers. Some samples, from the same site, were pooled together or cut in half as needed to acheive 10-30 grams of sample. Approximately 2 mls of BFM solution for every gram of sample weight was added to each beaker. A blank consisting of 50 mls of BFM solution was run along with every 4-8 samples to detect any contamination that may have been present.

The samples were digested by heating them over a steam bath for 4-6 hours.

The volume of each sample was then doubled with distilled water and placed in separatory funnels. Each sample was extracted 3 times using a total of approximately 90 mls of pesticide quality hexane and then poured off into a graduated mixing cylinder and wall shaken. After cooling, the total amount of hexane was then recorded. For lipid determination, 15-20 mls of the extract was then poured into a previously weighed beaker. The beaker was then re-weighed and the difference used to calculate the lipid weight of the sample. Approximately 10 mls of the extract was then placed into a graduated centrifuge tube and 1-2 mls of concentrated sulfuric acid was added and shaken for 1 minute, then centrifuged for 15 minutes. 5 mls of the cleaned-up extract was pipetted into a graduated concentrating tube and concentrated approximately 20 times using a roto evaporator and hot water bath.

(2) Sediment- Throughly frozen sediment samples were freeze dried for approx. 48 hours. A precise amount of dry sediment was weighed and mixed with sodium sulfate. This mixture was then transferred to a fritted glass extraction thimble and soxhlet extracted with approximately 500 mls of glass-distilled hexane for 12-18 hours. A blank consisting of a precise amount of sodium sulfate was run with every sediment series to detect any possible contamination of the sodium sulfate.

The hexane was then concentrated to approximately 25 mls on the roto evaporator and transferred to a graduated mixing cylinder. Total hexane was recorded and 10 mls of the extract was poured into a graduated centrifuge tube with 1 ml of concentrated sulfuric acid added. The centrifuge tube was shaken for 1 minute and then centrifuged for 10 minutes. The sulfuric acid was then frozen and the hexane poured into a vial containing 50-100 cleaned copper turnings. After approximately 12 hours, 5 mls of the cleaned-up hexane was pipetted into a graduated concentrating tube and concentrated 20-30

times before analysis by gas chromatography.

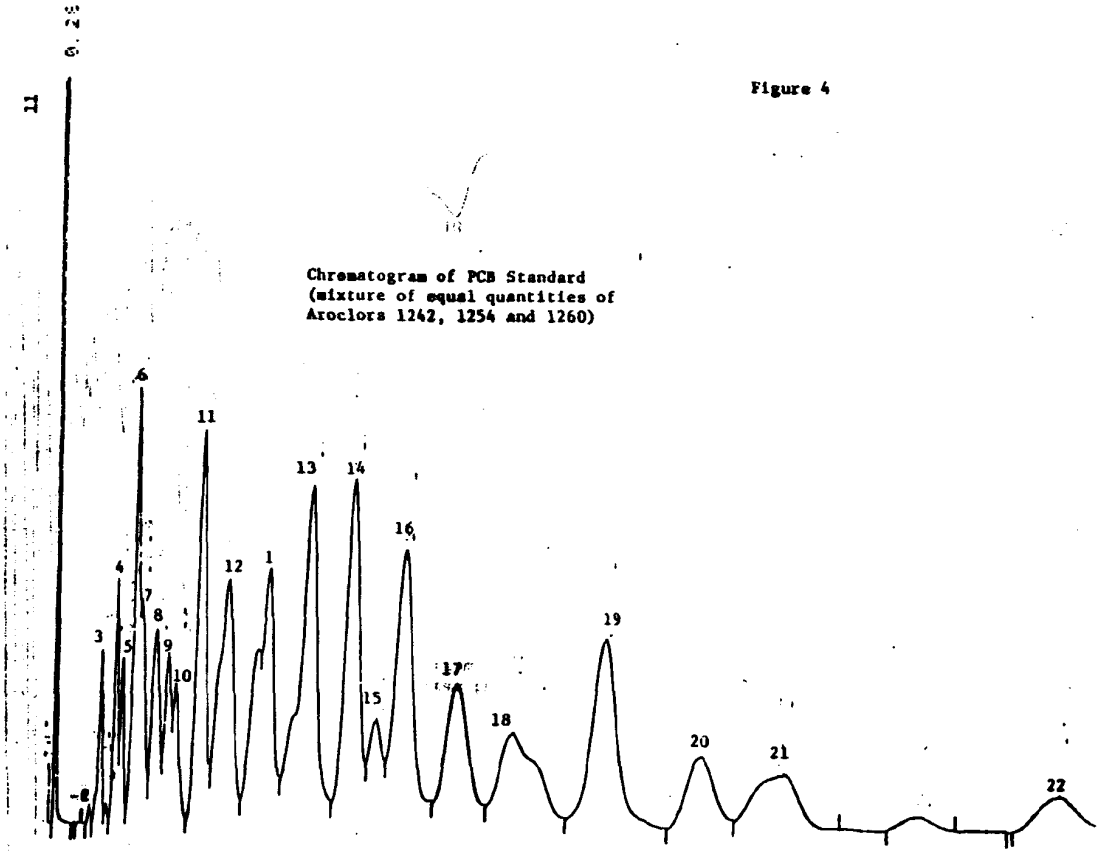
(3) Glassware Cleaning- All glassware used in sample analysis was cleaned by washing in hot soapy water followed by tap water rinse, acid rinse, tap water rinse, distilled water rinse, dry, pesticide quality acetone rinse, dry and pesticide quality hexane rinse. The glassware was then baked at 260°C for 12 hours, then capped with baked aluminum foil.

(4) Sample Analysis- A Hewlett-Packard 5100A electron capture gas chromatograph with ^{63}Ni detector on line with a Hewlett-Packard 3380A integrator or Perkin Elmer Pep-I GC data system was used to quantitatively determine the amount of PCB in each sample. A 6' coiled glass column was packed with a 1 in. alkaline pre-column containing 33% NaOH/KOH on Gas Chrom Q 80/100 mesh followed by 10% DC-200 on Gas Chrom Q 80/100 mesh. Instrument operating conditions were: oven temperature 210°C; detector temperature 300°C; carrier gas 95% Argon/5% methane at 60 ml/minute.

The PCB standard was prepared by mixing equal quantities of Monsanto Aroclor 1242, 1256 and 1260 in enough pesticide quality 2,2,4-trimethylpentane to yield 1.2 ng/ul concentration.

(5) Quantification- The amount of PCB in our samples was determined by identifying and quantifying the individual PCB homologs. In our PCB standard, 22 homologs and homolog pairs were identified by comparing their relative retention times and peak profiles with those published by Webb and McCall (1973). Figure 2 shows a chromatograph of our PCB standard with the peaks numbered. Figures 3 and 4 show a sediment and cottid sample respectively. A list of individual homolog and homolog pairs that are in our PCB standard is shown in appendix A. Identification of homologs in our samples was done by comparing relative retention times and peak profiles with those of our PCB standard. Concentrations of PCB in our samples were calculated by

Figure 4



Chromatogram of PCB Standard
(mixture of equal quantities of
Aroclors 1242, 1254 and 1260)

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Figure '3

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Chromatogram from Sediment Sample
from Site 1

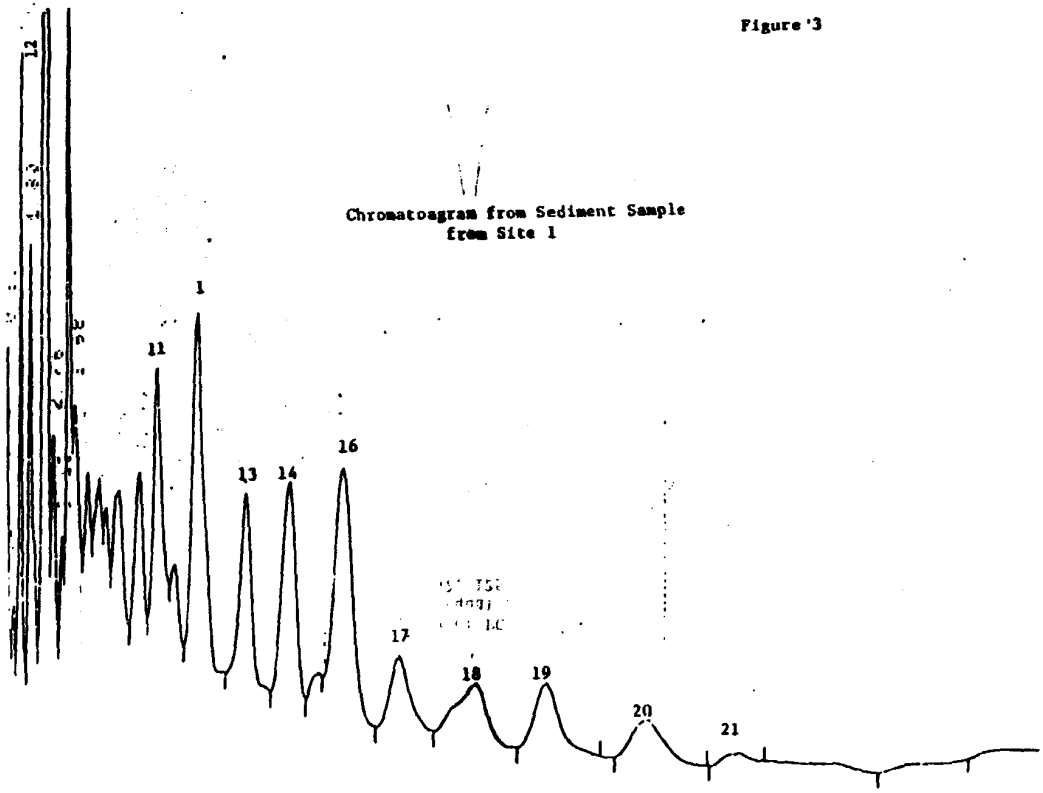
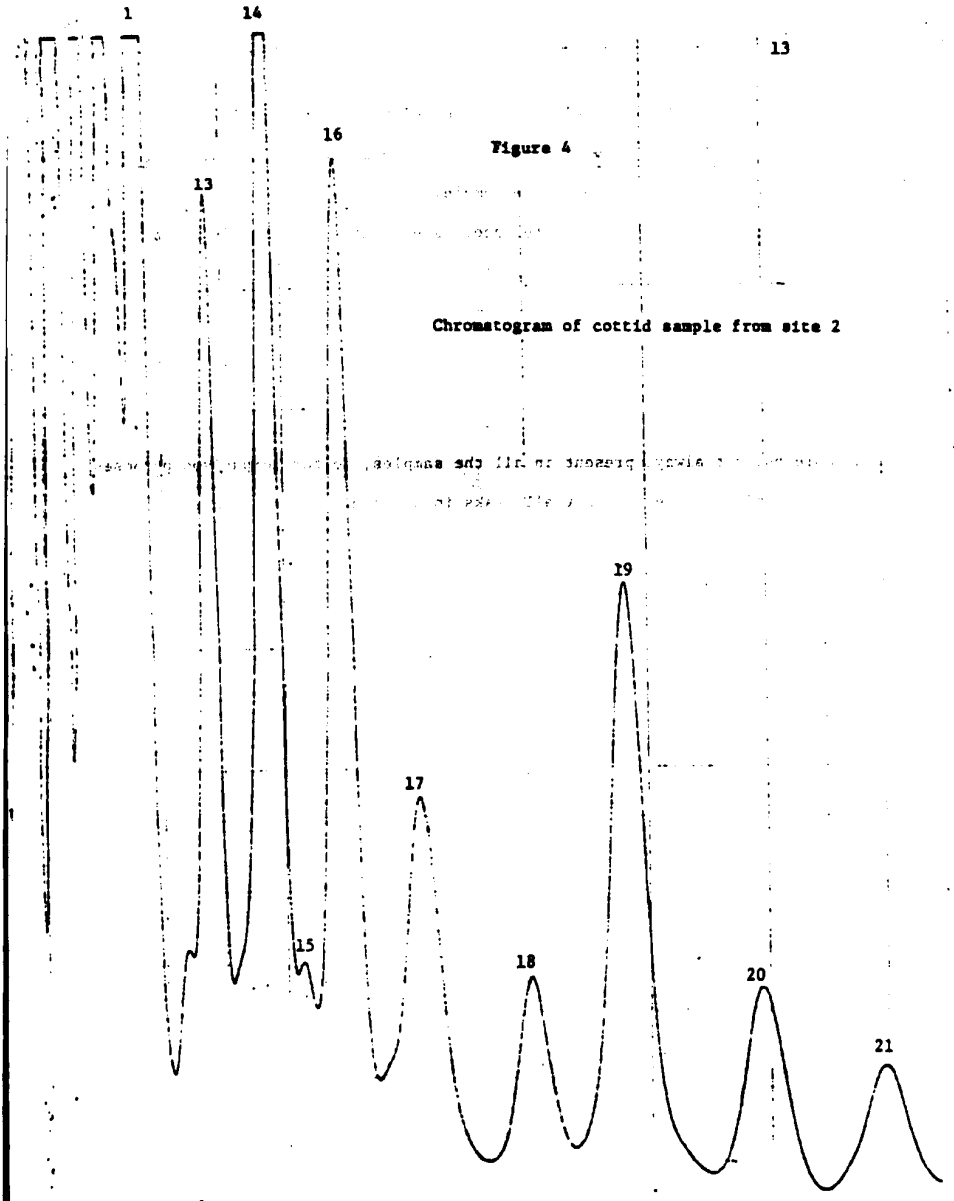


Figure 4

Chromatogram of cottid sample from site 2



quantifying each peak separately using mean weight percents published by Webb and McCall (1973). Nanograms and ppb wet weight were calculated for each identifiable peak and for the entire sample.

Peak 1, because of DDE interference, was estimated by establishing a relationship between peak 1 and surrounding peaks in the standard. We then applied the same relationship to our samples and arrived with an estimate of PCB and DDE for that peak. The concentration of PCB in our samples reflects the sums of peaks 11, 1, 13, 14, 17, 18, 19 and 20 (for cottid samples peaks 15 and 21 were quantified as well). Other peaks were quantifiable but not always present in all the samples, so for comparison purposes they were omitted. Because not all peaks in our samples were quantified our levels tend to reflect the minimum amount of PCB present.

RESULTS and DISCUSSION

The concentrations of PCB and DDE in the sediments and cottids of each of the 14 sites sampled on the Puyallup and White rivers are shown in table 1 and figures 5 and 6. Concentrations of PCB and DDE from the two dead western grebes found at site 1 are given in table 2. The highest levels in both cottids and sediments were found at sites 1 and 2. Both of these two sites are at the mouth of the Puyallup river which is heavily industrialized, having in it's proximity the Tacoma sewage treatment plant # 1, two chemical manufacturing plants and a St. Regis wood products mill. The sediment and cottid levels from sites 1 and 2 would tend to support the work of Mowrer et al. (1977) who reported a positive relationship between high PCB levels and heavy industrialization. The lowest level in sediments was found at site 7, although all sediment levels upstream from site 1 were less than 2 ppb (see figure 5). This relatively large drop in sediment levels from 10.2 ppb at site 1, to 2,0 ppb at site 2, was an unexpected finding. A possible explanation for this large difference in levels between two sites less than a 1/2 mile apart, could be that site 1 receives much of it's PCB contamination from Commencement Bay through the process of tidal washing of the river mouth. Support for this theory is provided by the fact that sediment levels similar to those we found at site 1 have also been reported by Mower et al. (1975) for sediments of Commencement Bay.

The lowest levels found in cottids were at sites 5, 6, and 10. With some exceptions, the highest levels in cottids at a site were found in the largest fish, with the reverse being true as well. In their study of Puget Sound, Mowrer et al. (1975) suggested that the levels they found in cottids were probably influenced by the levels in sediments, "as cottids have a tendency to

Table 1

CONCENTRATIONS OF PCB AND DDE IN THE SEDIMENTS AND CUTTIDS OF THE PUYALLUP AND WHITE RIVERS

Site No.	Sediments PCB (ppb)	DDE (ppb)	DDE/PCB	Cottids Sample Size	Wt. of sample (grams)	PCB (ppb)	DDE (ppb)	DDE/PCB
1	8.9	1.2	.13	2	28.1	227	35.5	.15
2	1.5	.40	.26	3	55.4	287.6	50.6	.17
3	1.2	.46	.38	-	-	-	-	-
4	1.1	.44	.40	5	21.1	129	16.6	.12
5	.70	.10	.14	2	9.4	55	13	.23
6	.80	.02	.025	1	17.9	16.3	8	.49
7	.70	.05	.07	1	16.8	132	37	.28
8	.11	.06	.54	-	-	-	-	-
9	.10	.04	.40	5	25	126	35.6	.28
10	.17	.04	.23	2	15.5	51	6	.11
11	.44	.17	.38	1	7.7	34	12	.35
12	.17	.06	.35	-	-	-	-	-
13	.15	.07	.46	-	-	-	-	-
14	.18	.16	.88	-	-	-	-	-

Figure 5

Concentrations of PCB in Cottids
from the Puyallup and White Rivers

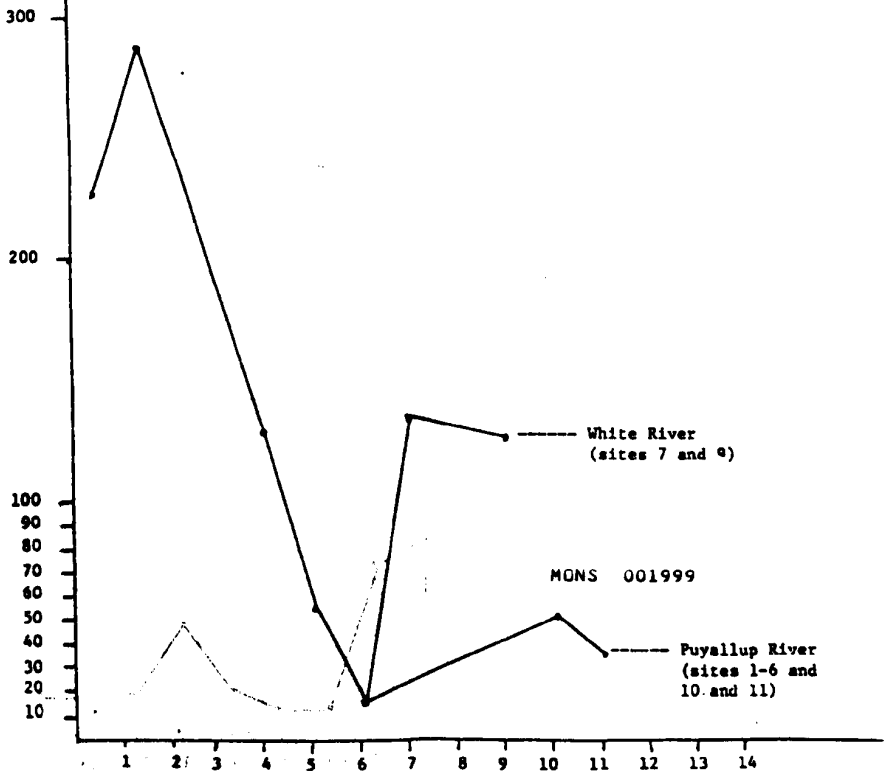


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Figure 6

Concentrations of PCB in Sediments
from the Puyallup and White Rivers

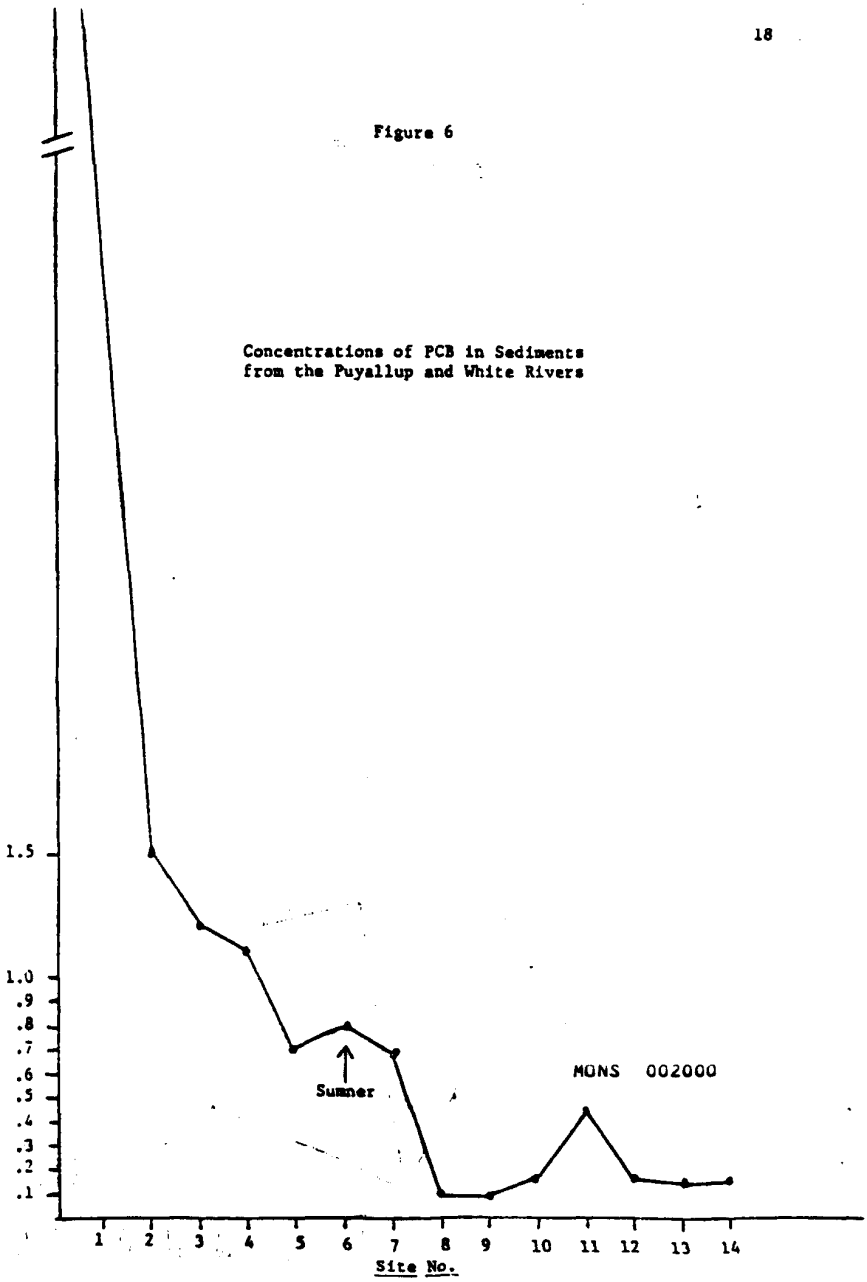

Table 2

CONCENTRATIONS OF PCB AND DDE IN TWO WESTERN GREBES FOUND DEAD AT SITE 1

	PCB		DDE	DDE/PCB
	Wet Wt. (ppm)	Lipid Wt. (ppm)	Wet Wt. (ppm)	Wet Wt.
Grebe 1				
Kidney	6.6	225.7	1.6	.24
Fat	405.2	476.6	86.6	.21
Grebe 2				
Liver	2.5	50	.83	.33
Fat	134	143	37.5	.27

forage for food in the sand or mud." The findings of our study would tend to support those of Mowrer et al., although we found much more variation between sediment and cottid levels than was reported by them. At site 1 for instance, the levels in cottids were roughly 25 times as concentrated as the levels in sediments, while at site 9 the levels between cottids and sediments differed by a factor of over a 1000. Fish have also been shown to accumulate PCB from suspended sediments (Zitko 1974). Since the Puyallup is a glacial river and tends to carry a large silt load, this kind of PCB accumulation could be as significant as direct accumulation from bottom sediments.

The third highest levels in cottids were found at sites 7 and 9 on the White (Stuck) river, the Puyallup's major tributary. This was an unexpected finding in that the PCB levels in cottids at site 6, three miles downstream from site 7 and six miles downstream from site 9, had only 1/8 as much PCB in their tissues as did cottids from sites 7 and 9. Possible explanations for these high levels could be the presence of the Fibre Board Industries plant at site 7, and the presence of the Lake Tapps power generating facility at site 9. The large drop in levels from sites 7 and 9 on the White (Stuck), to site 6 below the town of Sumner, is an example of how fast PCBs decrease with distance downstream from a source. In general, the levels found in the cottids of the White (Stuck) river (sites 7 and 9) are higher than those from other sites on the middle and upper Puyallup river (see figure 5).

In all samples both for sediments and cottids more PCB than DDE was present. DDE/PCB ratios ranged from .02 to .88 in sediments and from .11 to .40 in cottids. The ratios found in our study are similar to those reported

for sediments and fish in other large U.S. rivers (Henderson 1971).

Sediments and cottids sampled near sewage treatment plants (sites 2, 5 and 6) with the possible exception of site 2, did not show high levels of PCB as was initially expected. A more accurate measure of the impact of these sewage treatment plants on the Puyallup would be to directly monitor their effluent outfalls for PCB.

The levels found in the two dead western grebes collected at site 1 are difficult to interpret in terms of their biological and toxicological significance. The level found in the liver of one of the grebes (2.4 ppm) is much lower than levels found in the livers of Bengalese finches which had died from PCB poisoning (Dustman et al. 1971). However, another measure is that they are close to, or exceed altogether, the federal limit of 5 ppm PCB that is allowed in food fit for human consumption.

CONCLUSION

In conclusion, we found PCB to be a wide-spread contaminant throughout the 14 sites we sampled on the White and Puyallup rivers. From our data it would seem that the Puyallup river does not, at this time, have any large sources of PCB discharging into it's waters. It does appear, however, that in the areas of Fibre Board Industries and the Lake Tapps power generating facility,higher amounts of PCB are entering the river than at other sites upstream from site 1. The high levels found at site 1 are probably due more to industries that discharge into Commencement Bay, than from any upstream sources. The relatively high levels we found in the two dead western grebes is an example of how predators, through the process of biomagnification, can concentrate significantly higher levels of PCB than the prey they feed upon.

Although we conclude that PCB levels in the Puyallup and White rivers are low, we do not know whether or not they are biologically significant. Because PCBs have such a low water solubility and such a high sediment affinity, organisims in the Puyallup and White rivers will be exposed for many more years to constant, low level PCB contamination, the effects of which are still largely not understood by scientists.

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Appendix A

COMPOSITION OF PCB STANDARD

Peak No.	Mean Weight Percent	Homolog Name
2	0.97	1- chlorobiphenyl
3	3.77	2- chlorobiphenyl
4	3.67	2- & 3- chlorobiphenyl
5	2.03	3- chlorobiphenyl
6*	3.83	3- chlorobiphenyl
7*	3.70	3- chlorobiphenyl
8*	5.00	4- chlorobiphenyl
9*	3.23	3- & 4- chlorobiphenyl
10	2.33	4- chlorobiphenyl
11*	8.73	4- & 5- chlorobiphenyl
12*	9.43	4- & 5- chlorobiphenyl
1**	9.57	5- & 6- chlorobiphenyl
13*	10.73	5- & 6- chlorobiphenyl
14*	8.50	5- & 6- chlorobiphenyl
15	2.07	6- & 7- chlorobiphenyl
16*	6.93	6- chlorobiphenyl
17*	3.70	6- & 7- chlorobiphenyl
18	3.60	6- & 7- chlorobiphenyl
19*	3.67	7- chlorobiphenyl
20	1.40	7- chlorobiphenyl
21	1.33	8- chlorobiphenyl
22	0.50	8- chlorobiphenyl

*peaks used in quantifying our samples

**area of p,p' DDE overlap