Bull. Environ. Contam. Toxicol. (1986) 37:823–829
© 1986 Springer-Verlag New York Inc.

Environmental Contamination and Toxicology

Polychlorinated Biphenyl Residues in Some Marine Organisms from the Baie des Anglais (Baie-Comeau, Quebec, Saint-Lawrence Estuary)

C. Delval,* S. Fournier, and Y. Vigneault

Fisheries and Oceans, Fisheries Research Branch, 901, Cap diamant, P.O. Box 15500, Quebec, G1K7Y7, Canada

Polychlorinated biphenyls (PCB's) are among the most persistent compounds in the environment. These synthetic products are extremely resistant to biological and chemical degradation. In the high concentrations occurring by accumulation in the final links of the food chain, PCB's are poisonous for many organisms (Rubinstein et al. 1983). Their properties as electric insulators and their other industrial applications justified their usage until 1968, when their acute human toxicity was recognized during an incident in Yusho, Japan (Kuratsune 1972).

Although restrictions have been placed on their production in Canada since July 1980, limiting their use in capacitors and electrical transformers, PCB's continue to be released into the environment. During work involving the enlargement of harbour installations, studies conducted by an aluminum factory indicated levels of PCB's approaching 27 µg/g (dry weight) in sediments from the Baie des Anglais (68°06'30"W, 49°14'30"N) (Paul 1984).

The main source of PCB's in the Baie des Anglais comes from a pseudo lagoon which is located upstream from the Anse au Moulin immediately below the aluminum plant. Organic matter in suspension coming from industrial wastes from Baie-Comeau is likely responsible for the binding of PCB's. However due to hydrodynamic factors in the Baie des Anglais, contaminants stay only a short time in the littoral area and are evacuated to the southeast, where they accumulate (0.13 cm/year) at 80 m depth (Bertrand et al. 1985). Another source of contamination is the accumulation of dredging spoils at the entrance of the Anse au Moulin above the 40 m isobath. The contaminated sediments are resuspended by storm wave action (Bertrand et al. 1985) (Fig.1).

The work described in this paper was conducted in order to determine the extent of PCB bioaccumulation in two molluscs species (Mytilus edulis L. and Buccinum undatum L.) and two fish species (Clupea harengus harengus and Anguilla rostrata) from the Baie des Anglais (Baie-Comeau, Quebec).

---

*Correspondence and reprint requests. Present address: Station Marine, BP 41, 28,Avenue Foch, 62930 Wimereux France.

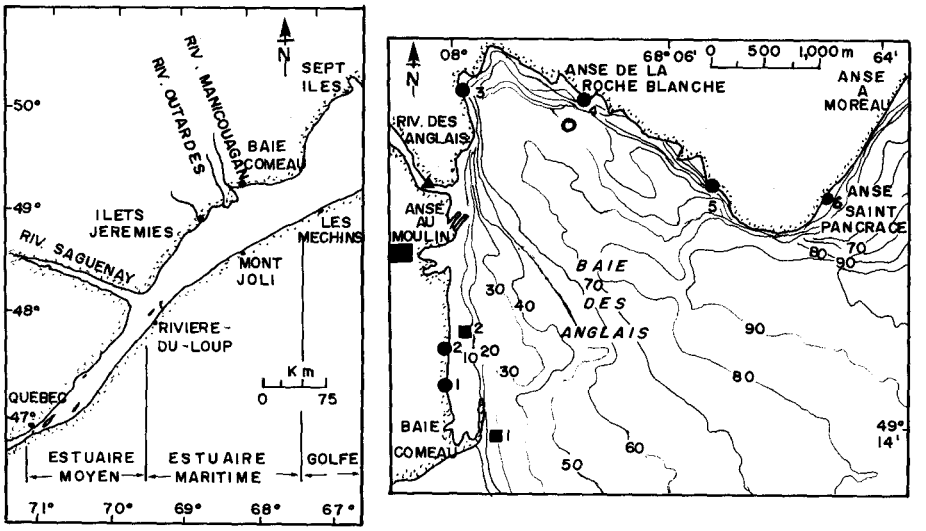

Figure 1. Localisation of sampling stations
(● :Mytilus edulis L.; ■ :Buccinum undatum L.;
o :Clupea harengus harengus; ▲ :Anguilla rostrata)

MATERIALS AND METHODS

Samples were collected during three periods, from May 30 to June 3, 1984, from June 24 to July 5, 1984 and from September 24 to 28, 1984. Mussels (Mytilus edulis L.), with a length between 31 and 46 mm, were collected from six stations in the Baie-Comeau area and from one control station in Ilets-Jérémie, 50 km southeast of Baie-Comeau (Fig.1). Whelks (Buccinum undatum L.), with a length between 57 and 70 mm, were taken from two stations by whelk traps from a commercial fishing boat. Due to their migratory behaviour, herring (Clupea harengus harengus) samples, obtained from a commercial catch, were available from only one station in the Baie des Anglais. The eels (Anguilla rostrata) were captured by eels pots and electric fishing from one site in the des Anglais river. The length of specimens, the number of samples and the PCB concentrations are summarized for each species in tables 1 to 3. The analyses were performed with 2 to 5 samples prepared as composites of specimens and the results were reported by considering the mean samples values and the standard deviation. The analyses were conducted on edible tissues, except for three whelks samples, which were separated into meat and viscera fractions.

The organisms were cleaned, wrapped in aluminum foil, and placed into polyethylene bags, then stored frozen at -4°C during transport. In the laboratory, the organisms were dissected, placed in glass jars and held at -20°C until analysis. The frozen samples were homogenized in an Osteriser Pulsematic blender just before analysis. The containers and aluminum foil under the lid were prerinsed with

hexane and acetone to prevent contamination. For analysis the samples were thawed and treated with sodium sulfate (Na_2SO_4 solid), extracted in an hexane-acetone mixture (4:1) by mixing, then concentrated at 38°C, under vacuum. The extractable organic compounds were then treated by the BEST method (Anonymous 1980), which consists of a chromatographic purification on a Florisil column (300 mm x 20mm i.d). The elution flow rate was 5 mL/min.

The identification and the quantitative analyses were conducted by Gas Liquid Chromatography (G.L.C). A Varian Model 3700 with a ^{63}Ni detector and an integrator (Varian Model 4270) were used. The column was a pyrex glass coil of 180 cm (i.d. 2 mm), packed with 4% OV-101 and 6%OV-210 on Gas Chrom Q of 80-100 mesh size for the initial identification, and 3% OV-101 on the same support for confirmation. Operating conditions were as follows: a)Temperature: injector, 258°C oven, 190°C; detector, 300°C; b)flow rate of nitrogen (high purity): 60 mL/min.

When the chromatogrammed compounds, such as PCB samples, must be compared to a standard or a standard mixture, quantification is preferably done using a computer and an integrator system. Since such a system was not available, we utilized the manual quantification method described by Reynolds and Cooper (1975). With this method, the total quantity of PCB's in a sample is determined by measuring the height of the peaks at 141 and 166, relative to the retention time of the pp'DDE load at 100, with a reference standard PCB mixture (Aroclor 1254:1260 - 1:1). The standards came from Polyscience Corporation. (6366 Gross Point Road, Niles, Illinois). The detection limit for PCB's was 0.002 µg/g for 10 g (wet weight) of initial homogenate. The precision was $\pm$ 2% for concentrations of 50, 100, and 200 µg/L of Aroclor standard. All determinations were done in duplicate and mean values and standard deviations of combined samples were calculated. The results are presented as concentrations (µg/g) on a wet weight basis for the whelks, herring, and eels. The mussels sample were freeze-dried before analysis, therefore the wet weight to dry weight ratio in these samples was determined and used to normalize the results (7.9 $\pm$ 1.9).

RESULTS AND DISCUSSION

The analytical results for PCB residues in the organisms studied are presented in tables 1 to 3.

The level of PCB's found in mussels from all seven stations were low and ranged from 0.002 to 0.046 µg/g wet weight (Table 1). The influence of PCB input from the Anse au Moulin was evident from the fact that samples from stations such as 1 and 2 tended to be 25 times higher in PCB concentration, than sample from station 5 (Saint Pancrace) and blank station (Ilets-Jérémie) which were near the detection limit. A decreasing gradient in the PCB concentration seemed to occur from the southwest (Baie-Comeau harbour area) (site 1) to the northeast (Saint Pancrace area) (site 6) (Fig.2). This seemed to be due to the hydrodynamic factors in the Baie des Anglais. The contaminants from the Anse au Moulin are evacuated to the south-southeast (Bertrand et al. 1985).

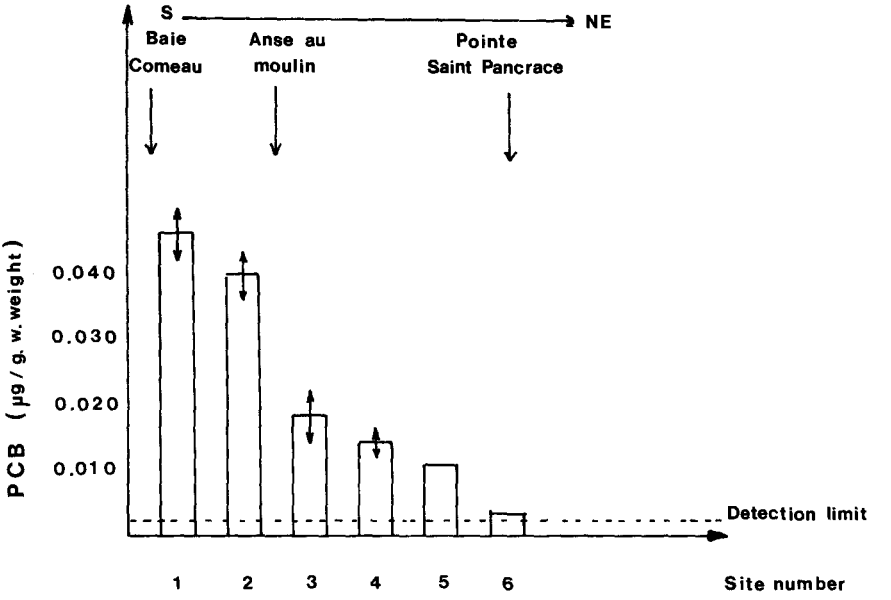

Figure 2. Concentration of PCB's in M.edulis L. from the Baie des Anglais.

However, the results from all seven stations are much lower than the 2 µg/g wet weight limit set by the Canadian Department of health and Welfare for PCB's in fish and shellfish. Cossa (1980), examined mussels from the St Lawrence estuary with a mean of 0.330 µg/g ± 0.14 µg/g dry weight. Our samples ranged from 0.364 ± 0.035 µg/g (dry weight) to 0.113 ± 0.017 µg/g (dry weight) for stations 1 to 4. These values were lower than those reported by Cossa et al. (1980) of 0.380 ± 0.380 µg/g (dry weight) for the corresponding area between Baie Sainte Catherine and Godbout. Moreover, they decreased with increasing distance from the principal waste source, at stations 5 and 6 (Fig.2).

Table 1. Polychlorinated biphenyls (PCB's) in M.edulis L. collected from various locations along the Baie des Anglais.

Site Number	Number of Specimens/ Sample	Average length (mm) $\bar{x} \pm$ sd	PCB µg/g (dry weight) $\bar{x} \pm$ sd	PCB* µg/g (wet weight) $\bar{x} \pm$ sd
1	103/2	37.61 ± 2.15	0.364 ± 0.035	0.046 ± 0.004
2	88/2	39.30 ± 4.69	0.320 ± 0.038	0.040 ± 0.004
3	121/2	33.54 ± 2.46	0.142 ± 0.046	0.018 ± 0.006
4	97/2	44.00 ± 2.92	0.113 ± 0.017	0.014 ± 0.002
5	51/1	33.14 ± 2.59	0.081	0.010
6	144/3	31.34 ± 2.55	≤ 0.024	≤ 0.004
Blank	103/2	35.55 ± 2.05	0.011 ± 0.002	≤ 0.002

* wet weight to dry weight ratio is 7.9 ± 1.9 s.d. :1

Levels of PCB's for whole whelk specimens ranged from 0.157 to 0.523 µg/g wet weight ($\bar{x}$= 0.292 ± 0.060 s.d. µg/g) at sample site 1 outside the public harbour, and from 0.125 to 1.00 µg/g (wet weight) ($\bar{x}$= 0.648 ± 0.40 s.d. µg/g) for sample site 2, near the west coast of the Baie des Anglais (Fig.1; Table 2). These concentrations are 5 to 10 times higher than the mean value found by Sloterdijk (1978) in whelks from the St Lawrence estuary (0.062 µg/g wet weight). Viscera, which contain an abundance of fatty tissues, are the preferred site for PCB's accumulation due to the greater solubility of these compounds in lipid reserves (Hutzinger et al. 1974). The level of PCB's in the viscera of whelks in our study was 500 times higher than in the meat (Table 2). Also, concentrations of PCB's in eviscerated specimens from station 1 was 150 times lower than that measured in the entire whelk. These results corroborate other data on the lipophilic behaviour of PCB's.

Table 2. Polychlorinated biphenyls in whelks (Buccinum undatum L.).

	Site Number	Number of Specimens/ Sample	Average length (mm) $\bar{x}$ ± s.d	PCB µg/g (wet weight) $\bar{x}$ ± s.d
Whole Whelk	1	40/4	63.06 ± 5.65	0.292 ± 0.06
	2	40/4	61.65 ± 6.24	0.648 ± 0.40
Meat	1	30/3	64.31 ± 5.17	0.002
Viscera	1	30/3	64.31 ± 5.17	0.925 ± 0.28

Concentrations of PCB's in whelks from the Baie des Anglais are high but do not exceed the federal limit of 2 µg/g wet weight recommended for the fish and shellfish industry. PCB levels in whelk were higher than those reported for mussels. This may be partly due to the higher relative proportion of lipids and to their respective positions in the trophic chain. In an estuarine area perhaps 80% of the PCB's from industrial wastes accumulate in the sediments (Pavlou et al. 1979), while the levels in the water column are comparatively low. Therefore mussels, which are filters feeding organisms are less susceptible to PCB accumulation than whelks. Whelks are sediments dwellers so they should be a better indicator for PCB contamination in the Baie des Anglais.

Table 3. Polychlorinated biphenyls in herring (C. harengus harengus) and eels (A. rostrata).

	Number of Specimens/ Sample	Average length (cm) $\bar{x}$ ± s.d	PCB µg/g (wet weight) $\bar{x}$ ± s.d
Herring	23 / 5	29.16 ± 1.79	0.068 ± 0.040
Eels	18 / 3	54.85 ± 8.4	1.13 ± 0.40

PCB levels in herring (C. harengus harengus) ranged from 0.007 to

0.139 µg/g wet weight ($\bar{x}$= 0.068 ± 0.04 s.d ug/g) (Table 3). These results are below those reported by Khalil et al. (1984) and Sloterdijk (1978) of 0.45 and 0.5 µg/g wet weight respectively. Different factors can affect the bioaccumulation of PCB's in herring. First, herring are migratory fish. Khalil et al. (1984) reported that the amount of PCB's is higher in individuals originating from spring populations than in those from autumn populations. In the St Lawrence estuary in july these populations are difficult to distinguish. So we were unable to determine the population source of our specimens. Second, the seasonal reproduction cycle can alter the metabolism of herring. For example, it is known that the annual reproductive cycle of herring is characterized by large fluctuations in the organism lipid content. It is difficult to compare our results with those of other researchers since we measured the PCB concentration in the meat and other studies are done in the entire fish, including their lipid reserves, eggs and gonads. For these reasons, herring is not good indicator of PCB contamination.

Eels (A. rostrata) are one of the most contaminated fish species in the Quebec area. Levels reaching 37.20 µg/g wet weight were measured in specimens from Lake Saint Pierre (Bélanger et al. 1977). For our study the mean concentration of PCB's in eels was high and exceeded by 12 times the low mean value found by others researchers in the estuary (about 0.10 µg/g wet weight)in the area between Tadoussac and Moisie which includes our study area (Desjardins et al. 1983). However, PCB concentrations in eels from the des Anglais River did not necesseraly reflect the contamination level in the bay. As Desjardins et al. (1983) reported, most of the eels in St Lawrence River come from its tributaries, and in particular, from Lake Ontario Nevertheless, it is perhaps significant that the PCB levels in eels from the Baie des Anglais exceeded those found in the atlantic tomcod (Microgadus tomcod), caught in the Hudson River estuary and at least 25% of the tomcod were attacked by cancer and other physiological perturbations (Klauda 1981).

Finally one can conclude, that of the species examined, the whelk is the best organism to monitor PCB pollution in the Baie des Anglais, in view of its distribution, nutritional mode and habitat. PCB concentrations in marine organisms from the Baie des Anglais are not alarming, but they are indicative of a generalized habitat deterioration. Further sampling is required to fully evaluate the contamination in the bay and to propose a restoration program.

Acknowledgments. This study was financially supported by the Ministry of Fisheries and Oceans (Canada). We are much indebted to Mrs Bastien, Mss Bertrand, Dubé, Schwab, and Verret for collecting samples and to Dr Dutil and his group for eel specimens. We also thank Mr Thibault, skipper of the Barbie-Joe , for his technical help at sea. We are grateful to Mr Walsh and Mr Sloterdijk for their comments about the manuscript.

REFERENCES

Anonymous (1980) Les méthodes d'analyse des pesticides organochlorés et des biphényles polychlorés. Bureau d'études sur les

Substances toxiques. Environment Quebec p 185
Bélanger C, Dubé JC, Lamontagne MP (1977) La présence de biphényles polychlorés dans l'environnement québecois. Pêches et environnement Canada. p 74
Bertrand P, Fournier S, Vigneault Y (1985) Concentration en biphényles polychlorés et en métaux dans les sédiments marins de la Baie des Anglais: contexte morpho-sédimentologique. Pêches et Océans Canada. Rapp Can Ind Sci Halieut Aquat (by press) p 112
Cossa D (1980) Utilisation de la moule bleue comme indicateur du niveau de pollution par les métaux lourds et les hydrocarbures dans l'estuaire du Saint Laurent. Pêches et Océans INRS-Océanologie Rimouski p 74
Desjardins C, Dutil JD, Gelinas R (1983) Contamination de l'anguille (Anguilla rostrata) du bassin du fleuve Saint Laurent par les biphényles polychlorés. Pêches et Océans. Rapp Can Ind Sci Halieut Aquat N°144 p55
Hutzinger O, Safe S, Zitko V (1974) The chemistry of PCB's. CRC Press, Cleveland. p 268
Khalil MZ, Labbe J, Lantagne S, Horth AC, Arnac M (1984) Les organochlorés dans l'estuaire du Saint Laurent. Sc et Techn de l'eau 17 1:73-75
Klauda RJ, Peck TH, Rice GK (1981) Accumulation of polychlorinated biphenyls in atlantic tomcod (Microgadus tomcod) collected from the Hudson River estuary, New York. Bull Environ Contam Toxicol 27:829-835
Kuratsune M (1972) An abstract of results of laboratory examination of patients with Yusho and of animals experiments. Environ Health Perspect 1:129-136
Paul M (1984) Baie des Anglais. Baie-Comeau. Synthèse des analyses de biphényles polychlorés dans les sédiments et la chaine alimentaire. Service de la qualité des eaux. Ministère de l'environnement du Québec. p 51
Pavlou SP, Dexter RN (1979) Distribution of polychlorinated biphényls (PCB's) in estuarine ecosystems. Testing the concept of equilibrium partitionning in the marine environment. J Mar Chem Soc 13:65-71
Reynolds LM, Cooper T (1975) Analysis of organochlorine residues in fish. Water Quality Parameters. ASTM, STP 573 S1:196-205
Rubinstein NI, Lores E, Gregory NR (1983) Accumulation of PCB's mercury and cadmium by Nereis virens, Mercenaria mercenaria and Palaemonetes pugio from contaminated harbour sediments. Aquatic toxicol 3:249-260
Sloterdijk H (1978) Rapport d'études sur le tronçon en aval de Montmagny. Contamination des organismes del'estuaire. Comité d'études sur le fleuve Saint Laurent. 9:709-819
Received December 23, 1985; Accepted January 31, 1986