Trends in PCB Contamination in Dutch Coastal and Inland Fishery Products 1972-1976

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INTRODUCTION

To the Dutch Ministry of Agriculture and Fisheries it is extremely important to have information concerning the contamination of fishery products with organochlorine pesticides and polychlorinated biphenyls (PCBs).

Earlier results (HAGEL and TUINSTRA 1974) from the Dutch fish monitoring program showed the presence of considerable amounts of PCBs to be a specific fishery problem. In contrast with most agricultural products of the Netherlands, in which at most traces of PCBs are found, the presence of PCBs in fishery products overshadows the total presence of other chlorinated pesticides. Therefore, fishery products may be considered as one of the main sources of PCBs in the average Dutch food.

The Dutch fish monitoring program is mainly directed to the most affected areas: the Yssel lake and the Dutch coastal waters. In this article we present the trends of PCBs contamination in these waters over the period 1972 - 1976.

SAMPLING

Commercial 10 kg fish samples were taken four times a year during the period January 1972 to the end of 1976 from the fishing grounds I - VI as indicated in figure 1. The edible part of sole, pike-perch, eel and mussel samples were freshly homogenized and stored deep frozen in glass jars until analysed.

METHODS OF ANALYSES

Twenty grams of the homogenized sample were grounded with anhydrous sodium sulphate and sand, and for 8 hours extracted with pentane. The pentane extractable material - the "fat" content - of the sample was determined by evaporating the solvent from part of the extract. Another part of the extract corresponding to 1 g of fish (for the eel corresponding to 50 mg of "fat") was used for the PCB determination.

Extraction and clean-up according to GREVE and GREVENSTUK (1975) were done only in the case of eel samples followed by silica gel separation of pesticides and PCBs (ARMOUR and BURKE 1970).

Gaschromatographic determination was carried out on a Varian 1740 apparatus with tritium electron capture detectors and a 3 % DEGS - 1 % phosphoric acid coating on a Gaschrom 100 - 120 mesh column, length 1.80 m, temperature 195 °C, injection volume 5 μ l.

QUANTIFICATION OF PCBs

It appeared that on the DEGS/ $\rm H_3PO_h$ column the peak in Arochlor 1254 with relative retention time of 1.24 to p,p'-DDE (= 1.00) had the same peakheight as the corresponding peak in Arochlor 1260. The same holds for the peak with relative retention time 1.69. These two peaks in the chromatograms were used for quantification with Arochlor 1254 as standard and the results averaged.

RESULTS AND DISCUSSION

In table I the PCBs content in the edible part of Dutch coastal and inland fishery products in the period 1972 - 1976 is given on a product and on a fat base together with the "fat" percentage of the different species investigated.

Because of their low solubility in water and high solubility in fat the majority of the PCBs is concentrated in the fat tissue of organisms. The ratio between the concentration of PCBs in water and in the fat tissue of organisms in a certain area will after some time reach a more or less constant value (HARVEY et al. 1974), where the fattiest fish shows the highest content on a product base. Therefore, comparable results independent of the original material may be obtained by expressing the quantities of PCBs in fish on a "fat" base. For example, there is a big difference in the mean PCBs content of pike-perch and eel of the Yssel lake on a product base (0.25 - 2.50 mg/kg), while on a "fat" base the results are much more similar (31 - 17 mg/kg). Also the PCBs levels in the other species become remarkable similar in this way.

A statistical consideration of the results from table I shows no significant change in the PCBs contamination of fish on a fat base in the different sampling areas during 1972 - 1976. Therefore, the average PCBs levels in these areas could be graphically represented as shown in figure 1.

From the results it may be concluded that all fish in the Dutch coastal waters and in the Yssel lake will contain in the order of 20 ppm PCBs on a fat base.

According to the mechanism proposed by HARVEY et al. (1974), it may also be concluded from table I that the PCBs levels in the water of the investigated areas remained more or less unchanged during the 1972 - 1976 period.

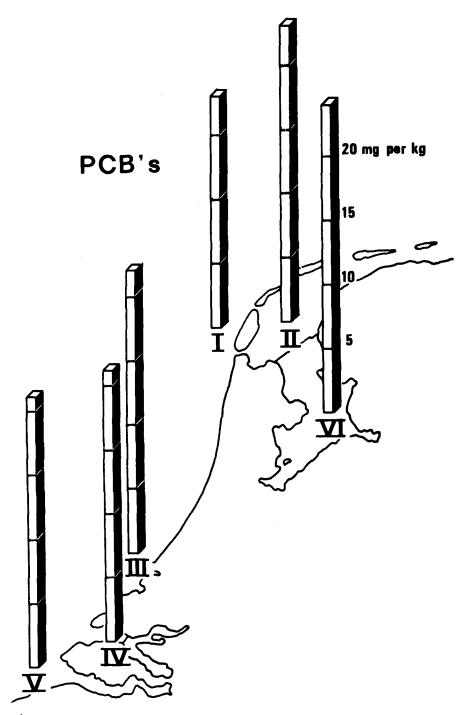
Table I

PCBs content in the edible part of Dutch coastal and inland fishery products 1972 - 1976, expressed as mg per kg on a product and on a fat base, together with the "fat" percentage of the different species.

Area	I			III			V			II			
fish		Sole			Sole			Sole			Mussel		
	Fat	PCB c tent (mg/k		Fat PCB content (mg/kg)		Fat	PCB con- tent (mg/kg)		Fat con-	PCB con- tent (mg/kg)			
Period	tent	pro- duct base	fat base	tent	1 -	fat base	tent %	pro- duct base	1	tent %	pro- duct base	fat base	
1972 - 1 2 3 4	2.6 1.9 1.3 2.8	0.23 0.23 0.27 0.18	9 12 21 6	1.0 1.7 1.7 1.8	0.14 0.52 0.79 0.43	14 31 46 24	1.6 2.1 2.1	0.26 0.77 0.55	16 37 26	1.8 1.8 0.9 0.4	0.33 0.26 0.16 0.18	18 14 18 45	
1973-1 2 3 4	2.7 1.2 0.9 1.1	0.21 0.21 0.14 0.07	8 18 16 6	2.3 1.1 2.2 1.5	0.25 0.33 0.49 0.17	11 30 22 11	0.8 3.3	0.23 0.49	29 15	1.1 1.5 0.9 0.8	0.29 0.21 0.20 0.15	26 14 22 19	
1974 - 1 2 3 4	0.6	0.28	47 19	2.0 0.8 2.4 2.7	0.12 0.25 0.80 0.63	6 31 33 23	1.2 0.6	0.12 0.29 0.12	10 48 9	0.8 0.9 0.3 0.7	0.15 0.12 0.07 0.13	19 13 23 19	
1975-1 2 3 4	2.1 0.4 1.1	0.38 0.09 0.10 0.25	18 23 9	2.0 0.6 2.3 1.7	0.32 0.24 0.64 0.16	16 40 28 9	0.3 0.9 2.5	0.12 0.13 0.39		0.4 1.2 0.7 0.9	0.20 0.23 0.14 0.24	50 19 20 27	
1976-1 2 3 4	0.6 1.1 3.5	0.32 0.23 0.45	53 21 13	1.2 1.7 1.2 3.8	0.12 0.29 0.27 0.39	10 17 22 10	0.9 1.0 2.2	0.19 0.12 0.21	21 12 10	0.4 1.3 0.9 0.9	0.14 0.14 0.14 0.21	35 11 16 23	

Table I (continued).

Area	·IV				ΨI		·VI			
fish		Mussel		P:	ike pe	erch	Eel			
	Fat PCB content con- (mg/kg)		Fat con-	PCB o tent (mg/k		Fat con-	PCB con- tent (mg/kg)			
Period	tent %		fat base	tent	pro- duct base	fat base	tent %	pro- duct base		
1972-1 2 3 4	3.0 2.5 1.3 1.0	0.40 0.33 0.20 0.23	13 13 15 23	1.0	0.38	38 60	19.0 16.2	3.12 3.27	16 20	
1973-1 2 3 4	1.1 1.2 1.4 1.2	0.37 0.22 0.19 0.17	34 18 14 14	0.8	0.21	26 29	23.8	2.25	9.4	
1974-1 2 3 4	1.4 0.8 1.0 0.9	0.20 0.20 0.15 0.16	14 25 15 18	1.0 1.0 0.9	0.16 0.42 0.35	16 42 39	7.1	3.6 2.10 3.74	28 30 18	
1975-1 2 3 4	0.8 1.0 0.8 1.1	0.28 0.25 0.11 0.21	35 25 14 19	0.8 0.6 0.9	0.26 0.11 0.24	33 18 27	8.1	2.1 1.9 2.7	16 23 13	
1976-1 2 3 4	0.6 0.5 0.8 1.0	0.31 0.12 0.10 0.14	52 24 12 14	0.7 0.5 0.6 1.2	0.24 0.14 0.15 0.24	34 28 25 20	16.4 18.1 20.8	1.9 1.4 3.0	12 7.7 14	



 $\frac{\text{Figure 1}}{\text{Average PCBs levels in Dutch coastal and inland fishery products 1972 - 1976, expressed as mg per kg on a fat base.}$

Although the PCBs values of table I on a product base show a small but significant decrease during 1972 - 1976, this can be totally accounted for by a simultaneous decrease in the fat content in some of the species investigated.

Finally, it can be seen from table I that all PCBs levels observed in the edible part of Dutch coastal and inland fishery products, expressed on a product base, are well below the tolerance level of 5 mg per kg suggested by the U.S. Food and Drug Administration. All samples, except eel of the Yssel lake, are on a product base also below a 1 mg per kg level.

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