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CONCENTRATIONS OF CHLORINATED HYDROCARBONS IN PIKE FROM THE TURKU ARCHIPELAGO IN THE YEARS 1970–1978

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Chlorinated hydrocarbons have been analysed from pike collected in 1970, 1971 and 1978 from 12 different sites in the Turku archipelago in Southwest Finland. The results are compared to values reported for pike in other parts of the Baltic. Concentrations of ΣDDT in pike decreased in the Turku archipelago during the 1970's, whereas concentrations of PCB compounds did not. None of the samples contained dieldrin and only small amounts of lindane, aldrin and hexachlorobenzene were found.

Index words: Pike, chlorinated hydrocarbons, PCB, DDE, DDD, DDT, lindane, aldrin, dieldrin, hexachlorobenzene, Baltic Sea.

1. INTRODUCTION

The first investigations of DDT and PCB concentrations in fish along the Finnish coast were carried out in 1969–1970 (Karppanen and Henriksson 1971, Karppanen et al. 1972). Hattula (1973) examined concentrations in pike, perch and bream offshore Helsinki in 1971 and Linko et al. (1974) in pike and Baltic herring from the Turku archipelago in 1972–1973. In the Baltic Baseline Study in 1974 (ICES 1977) concentrations of chlorinated hydrocarbons in Baltic herring, sprat, cod and plaice throughout the whole Finnish coastal region were investigated.

Chlorinated hydrocarbons were analysed from both freshwater and brackish water pike populations in the years 1970–1977 in conjunction with an investigation of mercury concentrations in pike which was started in 1970 by the Finnish National Board of Waters (Miettinen 1974). Pike was chosen because this fish species is the best indicator of residues in certain areas. Investigations were continued in 1978 in conjunction with a project for monitoring residues in freshwater and brackish water fish.

When considerable pollution of the Baltic Sea by DDT and PCB's was observed, the use of these compounds was subjected to control measures, although part of the pollution was shown to result from atmospheric transport from sites of utilization all over the world. As a result of these limitations the DDT levels of biota in the Baltic Sea were estimated to have passed their peak and begun to decrease in the latter part of the 1970's

(Olsson 1978). A similar decrease in levels of PCB compounds was not however observed. On the contrary concentrations of PCB compounds were estimated still to be increasing, mainly by atmospheric fallout (Kihlström and Berglung 1978). The aim of the present work was to determine the content of these compounds in pike from Turku archipelago during the period 1970–1978.

2. SAMPLING AND ANALYTICAL METHODS

Samples were collected in May 1970, 1971 and 1978. The location of the sampling sites is presented in Fig. 1. Each fish was first wrapped in aluminium foil and then closed into polyethene bags for preservation. Samples were stored deep-frozen at – 20 °C. Samples were partially thawed once before dissertation of the lateral muscle for the analysis of the chlorinated hydrocarbons. Pikes caught in 1970 were analysed in 1971 at the Department of Chemistry, University of Jyväskylä. Pikes collected in 1971 and 1978 were analysed by the Research Laboratory of the Water Research Institute in 1976 and 1978.

The analytical methods used by the Department of Chemistry, University of Jyväskylä have been presented by Hattula et al. (1978). The methods used by the Water Research Institute were as follows.

After drying with sodium sulphate, fat was extracted from biological material in a soxhlet, using a mixture of *n*-hexane, acetone, diethyl ether and petroleum ether. The fat was weighed after evaporation of the solvent. The content of organochlorine compounds in fat was analysed after treatment with aluminium oxide, sulphuric acid and chromic acid or alcoholic potassium hydroxide using a gas chromatograph equipped with a Ni^{63} electron capture detector and an analytical glass column containing 5.2 % QF-1 and 1.4 % SF-96 on Chromosorb W.

The standard for comparison contained a mixture of 10 ng/ml hexachlorobenzene (HCB), lindane (γ-BHC) and aldrin (HHDN) and 20 ng/ml *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT and

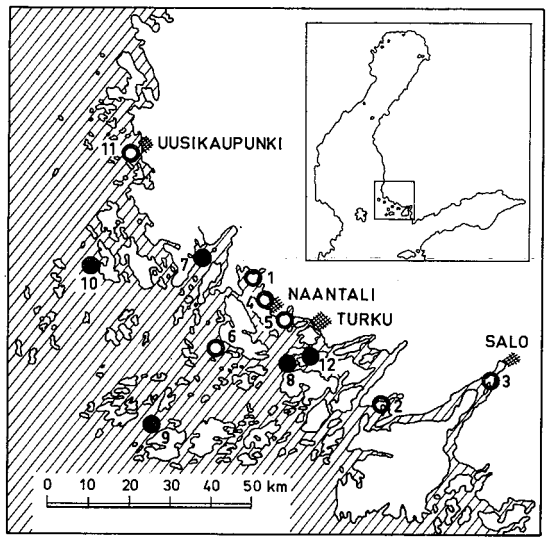

Fig. 1. Sites of pike sampling (○ 1970–1971, • 1978).

dieldrin (HEOD). The PCB standard used was Clophen A 60 for freshwater pikes (sites 1, 2, 3, 12) and Clophen A 50 for pikes from brackish waters. The concentrations of the PCB standards were 200 ng/ml.

The results were calculated by comparison of the heights of the corresponding peaks with the heights of the peaks of the standard solutions. HCB, γ-BHC and HHDN were calculated from the chromatograms of the fats treated with aluminium oxide. HEOD was calculated using the difference between the chromatograms of the aluminium oxide and sulphuric acid treated fats. DDT and DDD were calculated from the differences between the chromatograms of fats treated with sulphuric acid and with alkali. DDE was calculated using the difference between the chromatograms of the sulphuric acid and dichromate treated fats. The quantity of PCB's was estimated on the basis of all peaks detected (14 peaks). The heights of PCB peaks in the sample were estimated from the chromatograms after sulphuric acid treatment. The peaks interfered by DDE and DDD were measured from the chromatograms after chromic acid and alkali treatment.

The reliability of the data was assured in connection with the Baltic Baseline Study (ICES 1977). The results of the intercalibration

of the maize oil standards for the DDT- and PCB-type compounds lay around ± 20 % of the true value.

3. RESULTS AND DISCUSSION

Mean values and ranges of DDT and PCB compounds in fresh muscle tissue and in extractable fat are displayed in Tables 1 and 2 and mean concentrations obtained for the different areas in Table 3. Mean ΣDDT and PCB concentrations are presented in Figs. 2 and 3. Sites 1–3 represent river mouths, while 4–6 are sites off Turku and 7–10 are sites in the archipelago. For comparison one site (11) in the sea area off Uusikaupunki and one (12) in an island lake were included.

None of the 1971 samples contained dieldrin. Small amounts of lindane, aldrin and hexachlorobenzene were found in most samples in 1971. Concentrations in fresh tissue were below 0.005 mg /kg and ranged in extractable fat for lindane and aldrin from not detectable to 0.38 mg/kg and 0.49 mg/kg respectively and for hexachlorobenzene from 0.065 to 0.25 mg/kg.

The correlations of ΣDDT/fat-% and PCB/fat-% calculated on fresh weight basis for the whole research material were highly significant. Concentrations in extractable fat did not correlate with fat content. ΣDDT, PCB and fat contents did not correlate with fish weight. ΣDDT and PCB were mutually correlated with high significance for the whole material and significantly for the brackish water areas.

Table 1. Concentrations of DDT and PCB compounds in the lateral muscle of pike (n = number of specimens).

Sampling site	n	Weight g	Mean values and ranges (mg/kg fresh weight)				
			p,p'-DDE	*p,p'*-DDD	*p,p'*-DDT	ΣDDT[a]	PCB
1 Lemu	2	820 (780–910)	0.008 (0.004–0.013)	n.d.	0.003 (0.001–0.005)	0.012 (0.009–0.015)	0.011 (0.007–0.015)
2 Karuna	1	920	0.008	n.d.	n.d.	0.009	0.007
3 Salo	1	1 320	0.014	n.d.	0.005	0.021	0.018
4 Naantali	5	860 (770–1 000)	0.030 (0.018–0.045)	0.003 (0.002–0.004)	0.004 (0.003–0.005)	0.040 (0.025–0.056)	0.11 (0.063–0.24)
5 Turku	5	850 (770–950)	0.026 (0.015–0.040)	0.004 (0.003–0.005)	0.008 (0.006–0.011)	0.040 (0.027–0.056)	0.14 (0.063–0.19)
6 Rymättylä	5	1 080 (920–1 320)	0.043 (0.035–0.072)	0.011 (0.005–0.030)	0.034 (0.021–0.078)	0.094 (0.065–0.19)	0.11 (0.085–0.18)
7 Taivassalo	3	730 (700–780)	0.019 (0.013–0.028)	0.002 (0.001–0.002)	0.004 (0.002–0.007)	0.026 (0.019–0.039)	0.072 (0.049–0.11)
8 Airisto	3	820 (740–920)	0.011 (0.006–0.015)	0.002 (<0.001–0.006)	0.001 (0.001–0.001)	0.014 (0.009–0.018)	0.047 (0.029–0.059)
9 Korppoo	3	1 450 (1 150–1 680)	0.009 (0.007–0.011)	0.002 (0.001–0.002)	0.005 (0.002–0.006)	0.018 (0.013–0.022)	0.057 (0.042–0.069)
10 Kustavi	4	880 (750–1 210)	0.012 (0.005–0.023)	0.002 (n.d.–0.005)	0.005 (0.002–0.011)	0.021 (0.009–0.042)	0.063 (0.029–0.12)
11 Uusikaupunki	5	1 240 (1 060–1 470)	0.022 (0.014–0.036)	0.002 (0.002–0.004)	0.009 (0.003–0.015)	0.036 (0.021–0.059)	0.14 (0.12–0.18)
12 Lake Kaks-kerranjärvi	4	1 120 (650–1 670)	0.005 (0.004–0.007)	<0.001 (<0.001–<0.001)	<0.001 (<0.001–<0.001)	0.007 (0.005–0.009)	0.029 (0.022–0.35)

[a] ΣDDT = *p,p'*-DDT + 1.11 (*p,p'*-DDE + *p,p'*-DDD)
n.d. = Not detected

Table 2. Consentrations of DDT and PCB compounds in the lateral muscle of pike (n = number of specimens).

Sampling site	n	Fat contents (%)	Mean values and ranges (mg/kg in extractable fat)				
			p,p'-DDE	*p,p'*-DDD	*p,p'*-DDT	ΣDDT[a]	PCB
1 Lemu	2	0.16 (0.14–0.18)	5.2 (3.1–7.4)	n.d.	1.9 (0.34–3.4)	7.7 (6.8–8.6)	6.9 (5.1–8.7)
2 Karuna	1	0.15	5.3	n.d.	n.d.	5.9	4.2
3 Salo	1	0.19	7.2	n.d.	2.7	11	9.4
4 Naantali	5	0.42 (0.34–0.47)	7.1 (5.2–10)	0.67 (0.51–0.98)	0.87 (0.66–1.1)	9.5 (7.3–13)	25 (14–54)
5 Turku	5	0.58 (0.48–0.68)	4.8 (2.3–8.1)	0.72 (0.45–0.87)	1.5 (0.90–2.3)	7.6 (3.9–12)	27 (9.4–40)
6 Rymättylä	5	0.62 (0.43–1.3)	7.4 (5.7–8.8)	1.6 (0.95–2.4)	5.3 (4.1–6.8)	15 (13–18)	20 (15–26)
7 Taivassalo	3	0.52 (0.42–0.59)	3.6 (2.4–4.7)	0.30 (0.27–0.36)	0.70 (0.42–1.2)	5.0 (3.5–6.7)	14 (8.9–19)
8 Airisto	3	0.30 (0.28–0.32)	3.7 (2.2–5.2)	0.90 (0.15–2.2)	0.37 (0.32–0.43)	5.6 (5.1–6.3)	16 (10–20)
9 Korppoo	3	0.41 (0.37–0.48)	2.2 (1.8–2.9)	0.46 (0.26–0.58)	1.2 (0.65–1.6)	4.2 (3.4–5.5)	14 (11–17)
10 Kustavi	4	0.55 (0.36–0.84)	2.0 (1.6–2.7)	0.29 (n.d.–0.59)	0.83 (0.62–1.3)	3.4 (2.4–4.9)	11 (9.0–14)
11 Uusikaupunki	5	0.52 (0.48–0.59)	4.3 (2.7–7.6)	0.43 (0.30–0.80)	1.7 (0.72–3.1)	6.9 (4.2–12)	27 (21–35)
12 Lake Kakskerranjärvi	4	0.37 (0.29–0.44)	1.4 (1.1–1.7)	0.17 (0.11–0.27)	0.17 (0.12–0.19)	1.9 (1.5–2.4)	8.2 (5.8–12)

[a] ΣDDT = *p,p'*-DDT + 1.11 (*p,p'*-DDE + *p,p'*-DDD)
n.d. = Not detected

Table 3. Mean concentrations of ΣDDT and PCB in pike for the different areas and years in south-western Finland.

Area	Year	No. of specimens	Fat contents (%)	Mean concentration (mg/kg)			
				in fresh tissue		in extractable fat	
				ΣDDT	PCB	ΣDDT	PCB
River mouths	1970	4	0.16	0.014	0.012	8.0	6.8
Off Turku	1971	15	0.54	0.058	0.12	11	24
Off Uusikaupunki	1971	5	0.52	0.036	0.14	6.9	27
Turku archipelago	1978	13	0.45	0.020	0.060	4.5	13
Lake Kakskerranjärvi	1978	4	0.37	0.007	0.029	1.9	8.2

Significant differences in ΣDDT and PCB concentrations between different areas and sampling years, as well as in fat contents between different areas, were observed when the results were examined by one-way analysis of variance. Using the T-test the ΣDDT concentrations in fresh muscle tissue in pike collected off Turku were almost significantly higher than the corresponding results from river mouth sites. Concentrations in extractable fat did not, however, differ. In the case of PCB the concentrations in pike collected off Turku were highly significantly greater in fresh muscle tissue and significantly greater in extractable fat than the concentrations observed

in fish from the river mouth sites (Figs. 2 and 3). Fat content was also significantly higher in fish taken off Turku than in those from the river mouths (Fig. 4), so part of the difference in ΣDDT and PCB concentrations in fresh muscle tissue was presumably a result of difference in fat content. Concentrations of ΣDDT and PCB in pike collected offshore from Uusikaupunki in 1971 did not differ from those in pike offshore from Turku.

Concentrations of ΣDDT and PCB both in fresh muscle tissue and in extractable fat were at least significantly smaller in pike from the Turku archipelago in 1978 than in pike taken off Turku in 1971. Fat contents did not differ significantly between the two years. The ΣDDT concentrations in pike from Lake Kakskerranjärvi in 1978 were the lowest of the whole research material, whereas the PCB concentrations in extractable fat were similar to those of river mouth fish in 1970, and PCB content in fresh muscle tissue was actually twice as high in the lake fish (Figs. 2 and 3). At least in the case of PCB's the concentrations in Lake Kakskerranjärvi were a result solely of airborne transport of pollutants.

The results indicated that as a result of local pollution and pollution in the entire Baltic, the PCB concentrations in pike at the beginning of the 1970's were considerably higher in fish offshore from Turku than in those in river mouth areas. Increase in ΣDDT concentrations towards

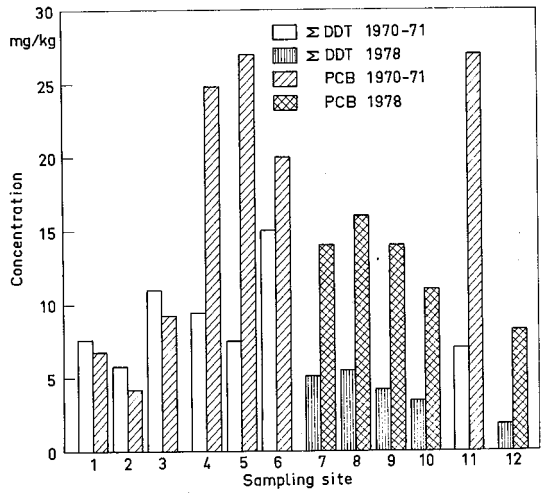

Fig. 3. Mean ΣDDT and PCB concentrations in the lateral muscle of pike (mg/kg in extractable fat) from different sites in the years 1970–1971 and 1978 in the Turku Archipelago.

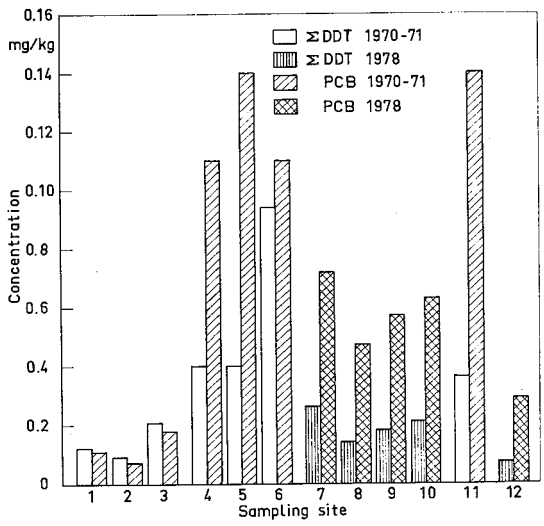

Fig. 2. Mean ΣDDT- and PCB concentrations in the lateral muscle of pike (mg/kg fresh weight) from different sites in the years 1970–1971 and 1978 in the Turku Archipelago.

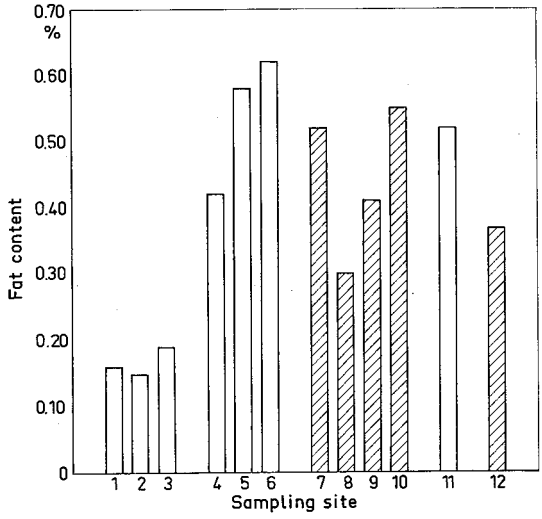

Fig. 4. Mean fat content in the lateral muscle of pike from different sites in the Turku Archipelago in the years 1970–1971 (unshaded columns) and 1978 (shaded columns).

the offshore areas was not so obvious, which probably reflects the earlier use of DDT in south-western Finland (Linko et al. 1974). Changes in concentrations during the course of this decade cannot be estimated solely on the basis of this research material, as the pike samples were taken from different sites in different years.

The differences in ΣDDT and PCB concentrations observed between pike taken off Turku in 1971 and those from further out in the Turku archipelago in 1978 may partly result from differences between the sampling sites or from different environmental conditions prevailing in the two years. For example in the spring 1972 Linko et al. (1974) found lower ΣDDT and PCB concentrations in the central area of the archipelago (Nauvo-Korppoo) than in either offshore from Turku or in sites further out in the archipelago. The ΣDDT concentrations found in 1978 in the present research were however considerably lower than those found by Linko et al. (1974) for the same areas:

			ΣDDT mg/kg	
		n	in fresh tissue	in extractable fat
Korppoo	1972	5	0.052	16
	1978	3	0.018	4.2
Kustavi-Taivassalo	1972	6	0.043	15
	1978	6	0.023	4.1

It would thus seem evident that ΣDDT concentrations in pike from the Turku archipelago have decreased during the 1970's.

The difference in concentrations of PCB-compounds between the 1971 and 1978 samplings in the present research, as opposed to the results for ΣDDT, probably resulted from differences between the sampling sites rather than between sampling dates. Comparison with the results of Linko et al. (1974) indicated that PCB concentrations had not decreased in the Turku archipelago during this decade:

			PCB mg/kg	
		n	in fresh tissue	in extractable fat
Korppoo	1972	5	0.039	13
	1978	3	0.057	14
Kustavi-Taivassalo	1972	6	0.053	19
	1978	6	0.068	13

ΣDDT and PCB concentrations observed by several authors at the beginning of the 1970's in pike from different areas of the Baltic Sea have been listed in Table 4. According to these investigations PCB concentrations along the Finnish and Swedish coasts were of the same order of magnitude. In the investigations carried out by the Finnish National Board of Waters the highest PCB concentrations were found off the coast near Turku, Uusikaupunki, Porvoo and Kotka (19–47 mg/kg in extractable fat). Concentrations in other coastal areas ranged from 5.3 to 16 mg/kg respectively. Karppanen and Henriksson (1971) also found high PCB concentrations (mean value 37 mg/kg in extractable fat) in the estuary of the River Kokemäenjoki. PCB concentrations along the Swedish coast in 1969–1971 ranged from 6.7 to 32 mg/kg respectively (Jensen et al. 1977).

According to the investigations by the Finnish National Board of Waters ΣDDT concentrations along the Finnish coast ranged from 2.1 to 15 mg/kg in extractable fat. The highest concentrations were found offshore from Turku and Porvoo. ΣDDT concentrations seemed to be somewhat lower than the concentrations observed by Jensen et al. (1977) in pike along the Swedish coast (4.9–28 mg/kg in extractable fat). According to Jensen et al. (1977) ΣDDT and PCB concentrations in pike were of the same magnitude throughout the Swedish coast of the Baltic Sea except for increased levels in some areas due to local sources of pollution.

In both 1971 and 1978 the ratio DDT/DDE increased with distance from the mainland and the ratio DDE/ΣDDT decreased (Table 5, Fig. 5). DDE represented in the river mouth and coastal sites 68–100 % and in the outer archipelago 54–66 % of all DDT compounds. Similar results

Table 4. ΣDDT and PCB concentrations in the lateral muscle of pike from different areas of the Baltic Sea at the beginning of 1970's compared with the results of the present study.

Area	Years	No. of specimens	Concentration (mg/kg)				Reference[b]
			in fresh tissue		in extractable fat		
			ΣDDT	PCB	ΣDDT	PCB	
Offshore Turku	1971	15	0.058	0.12	11	24	*1*
	1972	7	0.042	0.066	20	32	*2*
Turku archipelago	1972	39	0.039	0.048	15	21	*2*
	1978	13	0.020	0.060	4.5	13	*1*
Bothnian Bay	1969–70	15	0.008–0.011	0.026–0.055	2.3–4.2	11–19	*3*
	1971	18	0.019	0.046	4.3	11	*4*
	1969–71	66	0.064[a]	0.11	9.9	17	*5, 6*
Bothnian Sea	1969–70	9	0.013–0.031	0.021–0.134	4.0–9.2	5.0–37	*3*
	1970	7	0.016	0.031	6.2	11	*4*
	1969–71	191	0.043[a]	0.063	7.9	12	*5, 6*
Gulf of Finland	1969–70	10	0.026	0.035	12	17	*3*
	1970	10	0.015	0.028	6.0	13	*7*
	1971	37	0.051	0.15	8.0	26	*4*
Helsinki, Töölönlahti	1971	12	0.012	0.13			*8*
Offshore Stockholm	1969–71	30		0.11		20	*9*
Stockholm archipelago	1969–71	176		0.025–0.12		4.3–14	*9*
Baltic proper	1969–71	652	0.069[a]	0.067	14	14	*5, 6*

[a] Concentrations calculated from those reported by Jensen et al. (1977) in extractable fat

[b] 1 Present study
2 Linko et al. 1974
3 Karppanen and Henriksson 1971
4 Finnish National Board of Waters
5 Jensen et al. 1977
6 Kihlström and Berglund 1978
7 Karppanen et al. 1972
8 Hattula 1973
9 Olsson et al. 1973

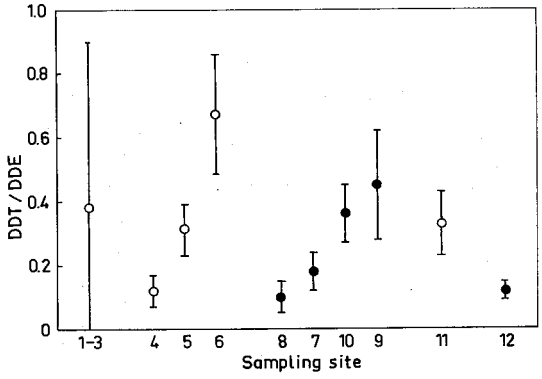

Fig. 5. Changing of the ratio DDT/DDE in the lateral muscle of pike from the Turku Archipelago (means ± standard deviations) in 1970–1971 (○) and 1978 (●). Distance from the connurbation of Turku - Naantali increases between sampling sites 4–6 and 8–9.

Table 5. Some ratios for the chlorinated hydrocarbons in pike. Distance from the connurbation of Turku-Naantali increases between sampling sites 4–6 and 8–9.

Sampling site	ΣDDT/PCB	DDE/ΣDDT	DDT/DDE
1	1.2	0.69	0.60
2	1.3	1.00	0.00
3	1.2	0.76	0.32
4	0.45	0.82	0.12
5	0.30	0.68	0.31
6	0.80	0.54	0.67
8	0.31	0.83	0.10
7	0.37	0.80	0.18
10	0.32	0.66	0.36
9	0.31	0.57	0.45
11	0.26	0.69	0.33
12	0.24	0.81	0.12

were observed previously for the Turku archipelago by Linko et al. (1974) and for the Stockholm archipelago by Olsson et al. (1973). Olsson (1978) suggested that this phenomenon could result from the capacity of the high biomass present in the inner archipelago, to decradate DDT into DDE and DDD, while the pelagic part of the Baltic is so strongly polluted by DDT that local influences do not notably increase the levels in biota. The fact that the ratio DDT/DDE was lower near open sea areas in 1978 than in the area of Rymättylä in 1971 (Fig. 5) indicates that decradation of DDT has taken place also in the pelagic part of the Baltic in the 1970's and not only in the Turku archipelago.

With the exception of pike from river mouth sites the ratio ΣDDT/PCB (Table 5) was in the present study lower than the average ratio of about 1 found by Jensen et al. (1977) in pike along the Swedish coast in 1969–1971. The ratio ΣDDT/PCB in pike from other Finnish coastal waters in 1970–1971 varied as follows:

Sea area	Range
Bothnian Bay	0.22–0.66
Bothnian Sea	0.51–0.67
Gulf of Finland	0.08–0.75

These values support the hypothesis that at the beginning of the 1970's the average ΣDDT concentrations in pike were lower in Finnish coastal waters than in Swedish coastal waters.

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Helsinki, September 1979

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LOPPUTIIVISTELMÄ

Vesihallituksen vuosina 1970–1971 suorittaman kalojen elohopeapitoisuustutkimuksen yhteydessä kerätyistä hauista analysoitiin DDT- ja PCB-yhdisteiden sekä lindaanin, aldriinin, dieldriinin ja heksaklooribentseenin pitoisuuksia Jyväskylän yliopiston kemian laitoksella ja vesihallituksen tutkimuslaboratoriossa. DDT- ja PCB-yhdisteiden pitoisuuksien tutkiminen kuuluu myös vesihallituksen vuonna 1978 aloittamaan sisä- ja rannikkovesien kalojen jäämäaineseurantaan. Tässä tutkimuksessa tarkastellaan kloorattujen hiilivetyjen pitoisuuksia ja niiden muutoksia Turun saariston hauissa sekä verrataan niitä muualla Itämeressä todettuihin pitoisuuksiin.

PCB-pitoisuudet olivat Turun edustan hauissa v. 1971 korkeita verrattuna Lounais-Suomen jokisuiden hauissa ja keskimäärin Itämeren rannikon hauissa mitattuihin pitoisuuksiin. ΣDDT-pitoisuudet noudattivat muun Suomen rannikon tasoa, vaikkakin korkein paikallinen keskiarvo oli Turun saaristossa. ΣDDT-pitoisuudet painottuivat 1970-luvun alussa Suomen rannikon hauissa pienempiin pitoisuuksiin kuin Ruotsin rannikolla mitatut. DDE muodosti jokisuissa ja rannikon tuntumassa 68–100 % ja ulompana saaristossa 54–66 % kaikista DDT-yhdisteistä.

ΣDDT-pitoisuudet olivat laskeneet Turun saariston hauissa 1970-luvulla. Sen sijaan PCB-pitoisuudet hauissa eivät olleet laskeneet alueilla, joilta oli tietoja vuosilta 1972 ja 1978.

Lindaani, aldriini, dieldriini ja heksaklooribentseeni analysoitiin vuonna 1971 pyydetyistä hauista. Yksikään näyte ei sisältänyt dieldriiniä. Lindaania, aldriinia ja heksaklooribentseeniä löydettiin lähes kaikista näytteistä, mutta pitoisuudet olivat tuorepainoa kohti laskien alle analyysimenetelmien alarajan.

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