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DDT and PCB residues in the arctic tern (Sterna paradisaea) nesting in the archipelago of southwestern Finland

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Arctic terns of different ages and their eggs were collected in the archipelago of SW Finland. The DDT and PCB residues of the samples were analysed by gas chromatography. In the adults there was a significant difference between the sexes, contamination being higher in the males, whose livers contained on average 31.4 mg/kg ΣDDT compounds and 80.1 mg/kg PCB compounds in extractable fat. In the females the amounts of these residues were significally smaller (21.4 mg/kg and 38.7 mg/kg, respectively). The reason for the difference is probably that laying females are able to shed part of their pesticide loads, including ΣDDT and especially PCB, into the eggs.

ΣDDT contamination of tern eggs was 8.1 mg/kg of lipid weight and that of newly hatched chicks 10.5 mg/kg, the corresponding values for PCBs being 27.7 mg/kg and 44.0 mg/kg, respectively. In chicks 2—3 weeks old, in contrast, contamination had decreased (mean ΣDDT 2.8 mg/kg and PCBs 10.8 mg/kg). For these reasons, when drawing up models of the accumulation of environmental pollutants along food chains, it is important to state the stages of the life cycle at which samples were taken.

No developmental defects in arctic terns could be attributed to chlorinated hydrocarbons during the productivity study in 1965—73.

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1. Introduction

Large amounts of environmental pollutants and simultaneously increased frequencies of development defects have been found in eggs and chicks of gulls and terns in the last few decades in some highly industrialized areas, especially on the coast of New York and on the Great Lakes, and in Alberta, Canada (Gochfeld 1971, 1975, Hays & Risebrough 1972, Switzer et al. 1971, 1973, Gilbertson 1975, Fox 1976). In the past chlorinated hydrocarbon pesticides caused great damage to tern populations on the coast of the Netherlands and Germany (Koeman & van Genderen 1972, Koeman 1975).

It is known that some environmental pollutants, especially PCBs, are found in large quantities in the organisms inhabiting the Baltic Sea (Jensen et al. 1969, 1972, Olsson et al. 1973). In the present work we analysed DDT and PCB residues in arctic terns (*Sterna paradisaea*) of different ages and in the eggs. The samples were collected in the archipelago of southwestern Finland in 1969—75. The arctic tern is a long-distance migrant, wintering on the coasts of Antarctica and South Africa (Salomonsen 1967, Saurola 1978). It is present in Finland only from the beginning of May to the end of July, and thus pollutants can accumulate in tern tissues anywhere between the Baltic and Antarctic. On the other hand, the amounts of environmental pollutants in the food chains of the Antarctic Sea are very small as compared with those in Baltic and the North Sea (Tatton & Ruzicka 1967, Brewerton 1969, Risebrough & Carmignani 1972). For testing the role of Baltic food chains in the process of contamination in arctic terns we also collected eggs in Lake Inari (69°N 28°E), northern Finland, where human agriculture and industrial activities are almost totally lacking.

2. Material and methods

2.1. Birds

Adult arctic terns were shot in the archipelago as follows: 30 May 1969: 1 female, 29 May 1970: 4 males, 2 June 1970: 5 females and 6 males, 1 July 1974: 2 females and 2 males. In addition, 19 arctic tern eggs and 27 chicks of various ages were collected from the same district in 1973—74. For comparison, ten eggs were sampled in Lake Inari, Lapland. The material was frozen and stored at —18°C.

DDT and PCB analyses were made from the pectoral muscles and livers of adult terns and older chicks. Only the liver tissues of newly hatched chicks were analysed, because an adequate sample was difficult to obtain from the pectoral muscles of small chicks.

The birds were thawed and the pectoral muscles and livers were removed. The material to be analysed was homogenized with a macerator and samples (2—5 g) were weighed. The eggs were thawed and homogenized with a macerator and samples (2—5 g) were taken for analysis.

The samples were homogenized again in a mortar with granular quartz, mixed with a fourfold amount of anhydrous sodium sulphate, dried overnight at room temperature and Soxhlet-extracted with a mixture of hexane, acetone, diethyl ether and petroleum ether (2.5:7.5:1:9 v/v) (Hattula 1973). The extracted fat was weighed and purified by TLC on Silica Gel (Linko et al. 1974).

2.2. Pesticide assay

TLC: The plates (20 × 20 cm) were covered with a layer of Silica Gel G 1 mm thick and heated at 200°C overnight. The absorbent on the plates was divided into three zones 5 cm wide, and 100 µml of sample solution (fat dissolved in methylene chloride, not more than 20 mg) was applied as a streak 3 cm above the lower edge of the plate. The plate was developed by a continuous ascending technique with an n-heptane-acetone mixture (99:1 v/v) until the solvent had migrated 15 cm above the starting line. After the plate had been dried at room temperature, the absorbent was divided into two sections above the fat front at 2.5 cm. The lower 5.0-cm section contained DDD and DDT and the upper 7.5-cm section DDE and PCBs. The size of each section was determined by using standard compounds and staining them with the silver nitrate reagent (Kovacs 1963). The adsorbent sections were scraped off and packed in chromatograph tubes (13 mm inside diameter), and the chlorinated hydrocarbons were eluted with a mixture of cyclohexane and diethyl ether (4:1 v/v). Eluate (2 ml) was collected and analysed by gas chromatography for identification and quantification of DDT and PCB residues.

Gas liquid chromatography: The analyses were conducted on a Varian Aerograph, gas chromatograph, Model 2440, equipped with two ^{3}H-electron capture detectors. The columns were 0.20 cm inner diameter and 183 cm long. The tubes were silanized and packed with 6 % (w/w) GE SF-96 on Chromosorb W.

In addition to the SF-96 column, a mixture of 1.5 % OV-17 and 1.95 % OV-210 (w/w) on Varaport was used to check the qualitative analysis. The operating conditions were as follows: the SF-96 and OV-17/210 columns were kept at 200°C. The flow rates of carrier gas (nitrogen) were 30 ml/min and 17 ml/min, respectively. In all analyses the temperature of the injection block was 210°C and that of the detector 220°C.

The standard compounds used were p,p′-DDT, p,p′-DDE, p,p′-DDD and Clophen A 60. The PCBs were quantified by summing the four highest peaks that did not interfere with the pesticide peaks and taking the mean value of them.

Recoveries of the above-mentioned compounds were determined for both eggs and pectoral muscle tissue of the arctic tern. The recovery percentages for p,p′-DDE, p,p′-DDD, p,p′-DDT and PCBs (Clophen A 60) were: pectoral muscle 86±4.4, 86±6.2, 86±7.8 and 73±12; eggs 101±14, 100±3.6, 89±14 and 104±2.1.

Owing to the wide variation in chlorinated hydrocarbon concentration in terns, statistical analyses were

Table 1. DDT and PCB residues (lipid and fresh weights, mg/kg, means and *SE*) in the livers and pectoral muscles of arctic terns of various ages on the basis of samples collected in the archipelago of SW Finland in 1969—74.

	Newly hatched chicks (*N* = 9)	Chicks aged 2—3 weeks (*N* = 19)		Adult ♀♀ (*N* = 9)		Adult ♂♂ (*N* = 11)	
	liver	liver	muscle	liver	muscle	liver	muscle
Fat-%	8.1±1.6	3.6±0.2	4.3	5.0±0.5	5.9±0.3	5.7±0.8	5.9±0.4
DDE							
lipid	7.1±1.6	2.1±0.4	2.6±0.3	17.0±8.9	21.0±10.7	27.5±5.8	26.9±7.7
fresh	0.7±0.2	0.1±0.01	0.1±0.02	0.7±0.2	1.0±0.4	1.6±0.4	1.4±0.4
DDD							
lipid	1.2±1.0	0.2±0.2	0.1±0.1	3.7±1.7	4.4±2.1	3.5±2.1	4.0±1.9
fresh	0.1±0.1	0.004±0.002	0.004±0.002	0.2±0.1	0.2±0.1	0.2±0.1	0.2±0.1
DDT							
lipid	2.1±0.5	0.04±0.02	0.2±0.1	0.4±0.2	0.8±0.4	0.3±0.3	0.7±0.4
fresh	0.2±0.1	0.01±0.004	0.01±0.003	0.02±0.01	0.1±0.02	0.02±0.02	0.04±0.02
ΣDDT							
lipid	10.5±2.3	2.2±0.4	3.0±0.5	21.0±10.5	26.1±12.8	31.4±7.4	31.5±9.6
fresh	1.0±0.3	0.1±0.01	0.1±0.02	0.8±0.3	1.3±0.5	1.8±0.5	1.7±0.5
PCB							
lipid	44.0±8.5	10.8±1.7	14.1±2.0	38.7±12.5	40.3±13.0	80.1±16.5	74.6±14.0
fresh	4.3±1.3	0.3±0.1	0.6±0.0	1.7±0.4	2.1±0.5	4.4±0.8	4.0±0.5

Table 2. DDT and PCB residues (lipid and fresh weights, mg/kg, mean and *SE*) in eggs of the arctic tern collected in the archipelago of SW Finland and Lake Inari, Finnish Lapland in 1974—75. (+ = traces).

	Archipelago ($N = 19$)	Lake Inari ($N = 10$)
Fat-%	10.8±0.9	12.1±1.5
DDE		
lipid	6.1±0.9	4.5±0.8
fresh	0.6±0.1	0.6±0.1
DDD		
lipid	0.4±0.1	+
fresh	+	+
DDT		
lipid	1.6±0.4	0.5±0.5
fresh	0.2±0.04	0.1±0.1
ΣDDT		
lipid	8.1±1.0	5.1±1.3
fresh	0.9±0.1	0.6±0.1
PCB		
lipid	27.7±3.9	23.6±4.6
fresh	2.9±0.4	2.8±0.6

made with a non-parametric test (Mann-Whitney one-tailed *t'* test). No annual differences in contamination levels inside the categories were found in the present material.

3. Results

3.1. Pollutant levels in relation to age and sex

The difference in ΣDDT and PCB residues between liver and pectoral muscle tissues was not statistically significant in either adults or 2- to 3-week-old young (cf. Särkkä et al. 1978).

Females secreted large amounts of chlorinated hydrocarbons into their eggs. Contamination of fresh tissues with PCBs was even heavier in eggs than in females (Tables 1 and 2). Larger amounts of PCBs were also found in newly hatched chicks, average contents being about 1.5 times those in eggs. But owing to the great variation amongst individuals the difference was not statistically significant (Table 3). The differences in ΣDDT residues were smaller, contents in eggs and newly hatched chicks being rather alike (Tables 1 and 2).

As the chicks grew, the concentrations of PCBs and ΣDDTs decreased. For instance, the liver lipids of the older chicks contained only 21 % of the ΣDDT and 24 % of the PCBs in those of newly hatched chicks. The difference was even greater in fresh liver (about 7—8 %). All the differences were highly significant (Table 3).

The chlorinated hydrocarbons were analysed in four chicks found dead in the nesting area (Table 4). Their ages ranged from some days to about one week. The ΣDDT residues in liver lipids were 1.8 times and PCB residues 2.3 times the respective levels in the killed chicks. In contrast, the concentrations in the fresh livers of the two groups were similar. The mean fat percentage was smaller in the chicks that were found dead (5.0 % versus 8.1 %), but individual variations were high (1.5—11.8 %).

No difference in contamination was observed between the females shot before and after the egg-laying period; however, the number of samples was small. Yet contamination was clearly less heavy in adult females than in males. PCB levels in females were only about half of

Table 3. Statistical significance of differences in ΣDDT (upper right corner) and PCB (lower left corner) levels in lipid and fresh tissues of eggs and livers between eggs, newly hatched chicks, chicks 2 to 3 weeks old, adult females and adult males of arctic terns. Symbols as follows: — = not calculated, ° = $P < 0.1$, * = $P < 0.05$, ** = $P < 0.01$, *** = $P < 0.001$, ns = not significant. Mann-Whitney one-tailed *t'* test.

		A		B		C		D		E	
		1	2	1	2	1	2	1	2	1	2
		ΣDDT									
A. Eggs											
1. lipid weights				ns	—	***	—	ns	—	—	—
2. fresh weights					ns	—	***	—	ns	—	—
B. Newly hatched chicks											
1. lipid weights		ns	—			***	—	ns	—	—	—
2. fresh weights		—	ns			—	***	—	ns	—	—
C. Chicks aged 2—3 weeks											
1. lipid weights	PCB	—	—	***				***	—	—	—
2. fresh weights		—	—	—	***			—	***	—	—
D. Adult ♀♀											
1. lipid weights		ns	—	ns	—	***			—	°	—
2. fresh weights		—	*	—	ns	—	***			—	*
E. Adult ♂♂											
1. lipid weights		—	—	—	—	—	—	***			—
2. fresh weights		—	—	—	—	—	—	—	*		

Table 4. Various metabolites of DDT and PCB residues (mg/kg) in the livers of young arctic terns found dead in the archipelago of SW Finland in 1974 (n = 4). (+ = traces).

	Lipid	Fresh weights
DDE	17.8±4.9	0.9±0.4
DDD	0.4±	0.1±
DDT	0.6±	+
ΣDDT	18.8±4.5	0.9±9.4
PCB	100.8±12.1	4.5±1.7

Table 5. Statistical significance of differences in ΣDDT (upper right corner) and PCB (lower left corner) levels in lipid and fresh tissues of pectoral muscles between chicks 2 to 3 weeks old, adult females and males of arctic terns. Symbols as in Tab. 3.

		A		B		C	
		1	2	1	2	1	2
				ΣDDT			
A. Chicks aged 2—3 weeks							
1. lipid weights				**	—	—	—
2. fresh weights		—		—	**	—	—
B. Adult ♀♀							
1. lipid weights	PCB	**			—	ns	—
2. fresh weights		—	***			—	ns
C. Adult ♂♂							
1. lipid weights		—	—	*	—		
2. fresh weights		—	—	—	**	—	

those found in males and although the difference in ΣDDT levels was less striking, the trend was parallel: the difference was either fairly significant or significant for livers (Table 3), but not significant in the case of ΣDDT contamination in pectoral muscles (Table 5).

3.2. DDE residues as percentages of the total DDT in terns

The percentage of ΣDDT present as DDE residues in terns varied both with the area and with the stage of the life cycle. In the eggs from the archipelago of SW Finland the proportion of DDE was highly significantly smaller than in the eggs from Lake Inari (Table 6, $P < 0.001$). The difference may have been caused by the higher proportion of DDE in the fish of Lake Inari. A few analyses made with small whitefish (*Coregonus lavaretus*) caught in Lake Inari indicate that they contain twice as much DDE, on a percentage basis, as does the three-spined stickleback (*Gasterosteus aculeatus*) in the archipelago of SW Finland.

Table 6. The percentage of various metabolites of DDT in the arctic tern eggs collected in the archipelago of SW Finland and Lake Inari (+ = traces).

	DDE	DDD	DDT	ΣDDT
SW archipelago				
lipid weights	76.0	4.5	19.5	100.0
fresh weights	74.1	4.7	20.0	98.8
Lake Inari				
lipid weights	89.3	+	10.5	99.8
fresh weights	91.8	+	8.2	100.0

Table 7. The percentages of various metabolites of DDT in livers and pectoral muscles of arctic terns of different ages.

	DDE		DDD		DDT	
	liver	muscle	liver	muscle	liver	muscle
Newly hatched chicks						
lipid weights	67.7	—	11.8	—	20.2	—
fresh weights	77.1	—	9.4	—	17.7	—
Chicks 2—3 weeks old						
lipid weights	95.0	88.1	7.8	3.1	1.8	6.4
fresh weights	85.7	91.7	5.7	3.3	7.1	7.5
Adult ♀♀						
lipid weights	81.0	80.2	17.4	16.7	2.0	3.0
fresh weights	87.5	79.2	21.3	16.9	2.5	3.8
Adult ♂♂						
lipid weights	87.6	85.3	11.2	12.6	1.0	2.3
fresh weights	88.0	84.4	12.0	12.6	1.1	1.2

In the latter area the percentage of DDE varied between about 80 and 95 % in older chicks and adult birds, but in the newly hatched chicks it was significally smaller and similar to the percentage in southern Finnish eggs (Table 6 and 7). Presumably embryos and young chicks are less efficient at metabolizing DDT to DDE than adults.

4. Discussion

As the ΣDDT and PCB levels are low in the organisms of the Antarctic (p. 141), it is likely that contamination of terns took place mainly along the migration routes on the west coasts of Europe and Africa and at the nesting places on the Baltic. The latter area is especially significant, because there contamination of organisms

has reached levels about tenfold those of the Atlantic and the North Sea (Jensen et al. 1969).

However, chlorinated hydrocarbons are so widespread that it is hard to determine the exact sources from which they have been accumulated. For instance, there was no significant difference in PCB contamination between samples of tern eggs collected in the SW archipelago and in Lake Inari. But the ΣDDT levels were significantly lower in the eggs from Lake Inari when expressed in relation to lipid weights ($P < 0.05$, Table 2). Analyses of the whitefish (*Coregonus lavaretus*) of Lake Inari showed that even in this lake the fishes contain DDT and PCB residues.

The females secrete pollutants into their eggs, and thus egg-laying tends to reduce the body load in the females (see also Särkkä et al. 1978). Moreover, it seems that PCB compounds are secreted more readily into eggs than DDT compounds. For instance, the PCB/ΣDDT ratio was 2.1 in the fresh liver tissue of the females, but 3.2 in the fresh tissue of eggs. The secretion of PCB is even more clearly indicated by the fact that the mean body load of PCB residues in male terns was over twice that in the females but the load of ΣDDT about 1.5. Perhaps the lipid composition of eggs is favourable for concentrating these chlorinated hydrocarbons.

Pollutant contaminations in the chicks were at their highest immediately after hatching and decreased with the growth of the chicks. Such observations have been made with shags (*Phalacrocorax aristotelis*), guillemots (*Uria lomvia*), herring gulls (*Larus argentatus*) and some anatids (Robinson et al. 1967, Jensen et al. 1970, Charnetski 1976, Teeple 1977). For this reason, embryos and newly hatched chicks are stages especially susceptible to the lethal effects of environmental pollutants.

Table 8 lists the DDE levels found in tern eggs in various parts of Europe and North America. The largest amounts have been found on the coast of New York and at Chip Lake, Alberta. In these localities high mortality and increased frequencies of developmental abnormalities were also found among chicks (Switzer et al. 1973, Hays & Risebrough 1972). The tern eggs in the archipelago of SW Finland contained the same amounts of DDE residues as those from the coasts of the North Sea. Although PCB contamination is rather high in the arctic terns nesting in the study area, it is small compared with that found in common terns in the Great Lakes of Canada (Vermeer & Peakall 1977).

No signs of developmental abnormalities or increased frequencies of egg and chick mortality were found among newly hatched chicks in the Finnish archipelago. During 1965—73 some 800 newly hatched chicks were kept under regular observation from hatching to the age of 14 days, but only one was abnormal, an eyeless chick with a crossed beak. The higher pollutant levels found in tissue lipids of dead chicks (p. 143) are probably due to the fact that the chicks were starved and the poisons concentrated into the remaining lipid.

Of the 832 eggs watched only 5.3 % failed to hatch owing to infertility or embryonic death. This percentage is clearly lower than the 53 % reported by Fox (1976) for common terns in Buffalo Lake or the 15.3 % and 17.6 % found by Pettingill (1939) and Hawksley (1957) in the arctic tern colonies of Canada in the 1930s and 1940s, before the industrial mass production of DDT and probably also PCB compounds. For these reasons, it is highly improbable that environmental pollutants were responsible for the embryonic deaths, abnormalities or mortality in chicks of the arctic terns nesting in the archipelago of SW Finland.

Table 8. Mean amounts of DDE in eggs (fresh weight, mg/kg) of various tern species from Europe and North America.

	N	DDE	Author
Sterna paradisaea			
Archipelago of SW Finland	(19)	0.63	Present study
Lake Inari, Lapland	(10)	0.56	—»— —»—
S. hirundo			
Chip Lake, Canada 1968	(10)	25.2	Vermeer & Reynolds 1970
—»— —»— 1969	(68)	7.6	Switzer & Lewin 1971
—»— —»—1970	(105)	4.5	—»— —»—
Montreal Lake, Canada 1968	(10)	15.9	Vermeer & Reynolds 1970
Primosa Lake, Canada	(10)	2.0	—»— —»—
New York, USA	(5)	3.6	Woodwell et al. 1967
Coquet Island, Great Britain	(5)	0.04	Robinson et al. 1967
S. sandvicensis			
Farne Island, Great Britain	(8)	0.75	Robinson et al. 1967
Holland 1970	(10)	0.4	Koeman et al. 1972

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