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ORGANOCHLORINE PESTICIDES AND POLYCHLORINATED BIPHENYLS IN FOODS IN SWEDEN

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A survey of investigations of organochlorine pesticides and PCB in foods during 1967—1972 is presented. Only occasionally did samples of vegetable foods exceed the Swedish tolerances for organochlorine pesticides. Of the animal products, fat fish from certain waters contained high levels of pesticides and PCB. The highest concentrations of DDT and metabolites were found in cod liver from the Baltic. Some samples of cereal products were contaminated with PCB from the packaging material. There is a manifest decrease in the level of dieldrin in fish from certain waters and of p,p'-DDT in human milk from the Stockholm region. The average level of PCB in the human milk has increased.

The investigations presented here were performed at the Swedish National Institute of Public Health and at the Swedish National Food Administration.

The determination of organochlorine pesticides in vegetable foods by gas chromatography with electron capture detector was started in the early sixties. This investigation has been continued and later extended to animal products. The identity of the pesticides found by gas chromatography was confirmed by thin-layer chromatography. The methods used for extraction, clean-up and final analysis have been described elsewhere (1, 2, 3, 4).

Investigations of non-fatty foods

The latest investigation of vegetable foods (5) comprises about 2500 samples from the period 1968 — June 1972 and includes fruits, berries, vegetables, juices, mushrooms, honey, and soya products. It includes products from 36 countries. About 900 samples (37%) are of domestic origin. Fresh, dried and canned products were analysed. Swedish tolerances for organochlorine pesticides have only been established for *fresh* fruits, berries, vegetables, roots, and potatoes (Table 1). In this investigation 1.2% of all the fresh fruits, berries and vegetables examined

Table 1
Swedish tolerances for organochlorine pesticides in or on fresh fruit, berries, vegetables, roots, and potatoes

Pesticide	Food	Tolerance mg/kg
Aldrin and dieldrin together	Carrots	0.05
	Other commodities concerned	0.1
DDT	Pears, apples	2
	Other commodities concerned	1
Lindane	All commodities concerned	1

exceeded the Swedish tolerances for organochlorine pesticides. Only imported products exceeded the tolerances (Table 2). Apples, pears, plums, grapes, and tomatoes exceeded the tolerance for DDT, cucumber exceeded the tolerance for dieldrin and lettuce the tolerance for lindane. One sample of dried currants exceeded the tolerance for DDT in fresh fruit.

Table 2
Products exceeding the Swedish tolerance for organochlorine pesticides

Imported product	Number of samples analysed	Number of samples exceeding tolerance
Apples	140	2
Pears	156	1
Plums	37	1
Grapes	145	10
Cucumber	139	3
Tomatoes	204	1
Lettuce	96	9

88% of the samples had pesticide levels of 1/10 of the tolerances at the most. In this investigation sampling was partly concentrated to products where high pesticide levels were expected.

Several pesticides for which there are no Swedish tolerances were found in relatively high concentrations. Quintozen (pentachloronitrobenzene) and its metabolite pentachloroaniline were found in domestic

carrots and parsley and in Dutch lettuce. In carrots the highest concentration was 3.3 mg quintozen/kg and in lettuce 3.5 mg quintozen/kg. For lettuce the international expert group of FAO/WHO has recommended a tolerance of 0.3 mg quintozen/kg.

Dicofol (Kelthane) was found in many samples of apples and pears. The highest level, 3.5 mg/kg, was found in apples from Argentina.

Tetradifon (Tedion), endosulfan (Thiodan) and chorofenson were also found, but in lower concentrations.

About 300 samples of cereal products (bread, biscuits, macaroni products, grains, flour, breakfast cereals, and children's foods) have been analysed (4). Lindane was found in most of these samples. The levels were usually below 0.1 mg/kg. However, one sample of imported breakfast cereals contained 1.5 mg lindane/kg, and several samples of crisp bread contained lindane above 0.5 mg/kg. In the samples of crisp bread with relatively high lindane levels we also noticed an unknown peak in the gas chromatogram. This peak probably represented a product formed by thermal decomposition of lindane. Heating of a solution of lindane in dimethylformamide resulted in a mixture of decomposition products, one of which gave identical retention times on gas chromatography and identical R_f-values on thin-layer chromatography with the unknown substance in the bread samples. The substance was analysed by IR and mass spectrometry and identified as γ-1,3,4,5,6-pentachlorocyclohexene.

Some of the cereal products contained PCB (polychlorinated biphenyls). As high a level as 11 mg PCB/kg was found in one sample of imported children's food, which had been stored at room temperature for 2 years. In this kind of children's food the PCB found was identical with Clophen A 60, as shown by gas chromatography (Fig. 1) and thin-layer chromatography (Fig. 2). The origin was the packaging material. The highest concentration of PCB was found in the coloured side of the cardboard box (Fig. 3). The sale of this children's food was prohibited in July 1971.

Other types of PCB (Clophen A 30 and Clophen A 40) were also found in cereal products and the origin was the packaging material (Fig. 4 and 5). In these cereal products the PCB level was 1 mg/kg at the most.

Investigations of fatty foods

In 1967—1969 about 1400 samples of margarine, vegetable oils and foods of animal origin were analysed (6). In Sweden there are no tolerances established for pesticides in these foods. In relation to the acceptable daily intakes of chlorinated pesticides and the magnitude of consumption, high levels of pesticides were found in some samples. However, most of the samples contained low levels of pesticides. 90% of the samples is this investigation contained at the most 0.1 mg/kg of the DDT group (DDT+DDE+DDD), lindane, α-BHC and PCB and at the most 0.01 mg/kg of dieldrin.

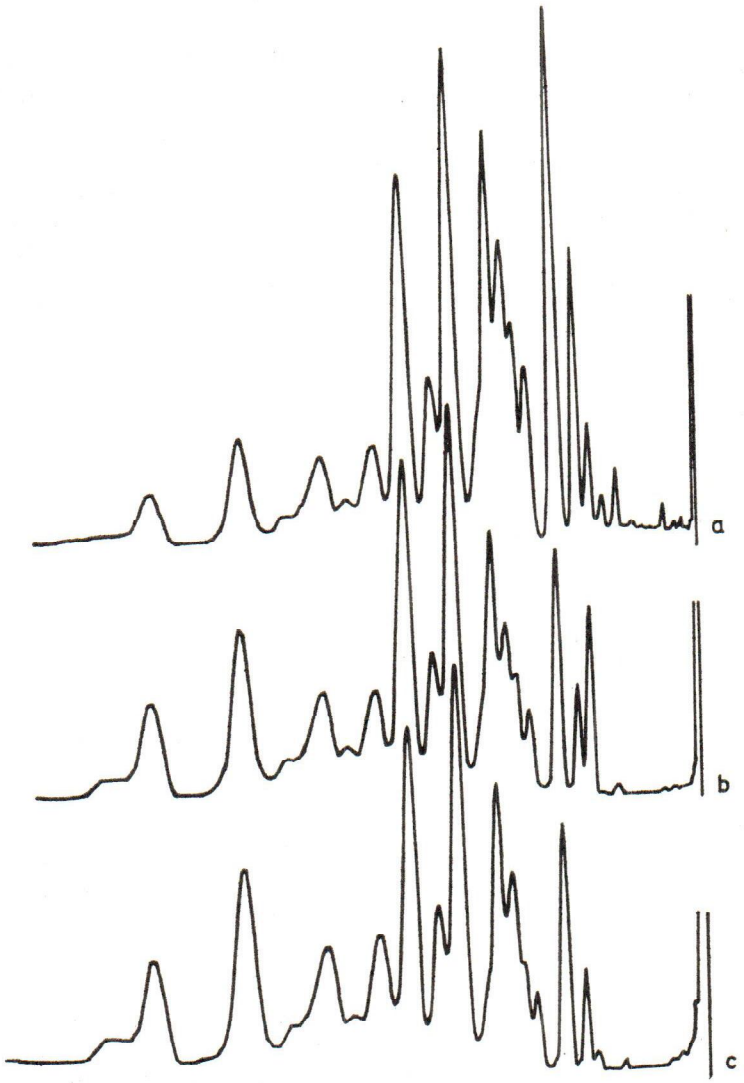

Fig. 1. *Gas chromatograms of a) PCB in a children's food, b) PCB in a cardboard box of this children's food, and c) PCB, Clophen A 60*

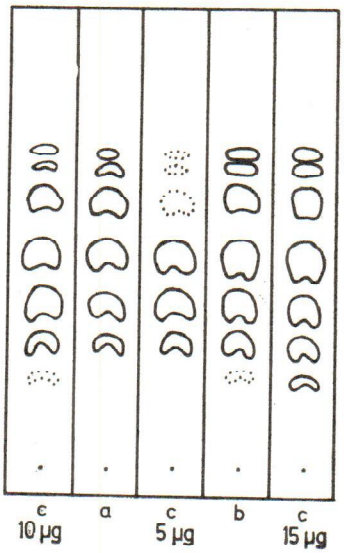

Fig. 2. *Thin-layer chromatograms of a) PCB from a children's food, b) PCB from the cardboard box of this children's food, and c) PCB, Clophen A 60*

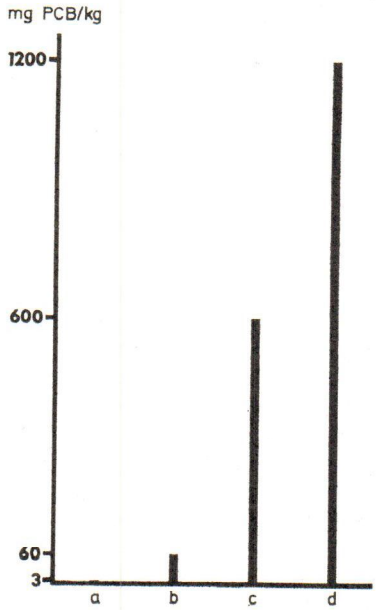

Fig. 3. *PCB levels in an imported children's food and amount of PCB extracted from different parts of its packaging material, mg/kg. Extraction for about 5 hours at room temperature a) children's food, b) inner bag, c) inner side of cardboard box, d) outer, coloured side of cardboard box*

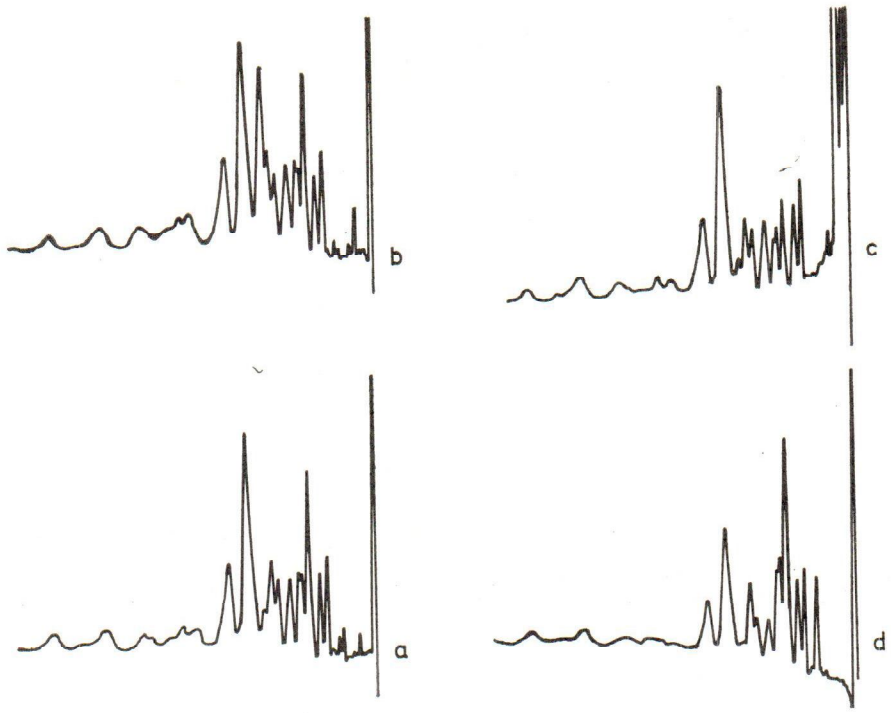

Fig. 4. *Gas chromatograms of PCB in breakfast cereal a) after clean-up thin-layer chromatography (3), b) after clean-up according to a) and boiling with sodium hydroxide solution (3) c) after clean-up according to a) and b) and oxidation with chromic acid (3) d) Gas chromatogram of PCB, Clophen A 30*

Margarine and vegetable oils contained very low levels of pesticides, with the exception of one sample of sesame oil (DDT 0.33 mg/kg, lindane 0.65 mg/kg, α-BHC 0.73 mg/kg, and dieldrin 0.02 mg/kg). The analysed foods of animal origin (cow's milk, butter, human milk, egg, baby food, and meat of poultry, pig, lamb, ox, and fish) usually contained pesticides, although in most of the samples the concentrations were low. In relation to the daily intakes, human milk had high levels of DDT, DDE and dieldrin. One sample of egg yolk contained 7.6 mg/kg of the DDT-group. Lindane was found in several samples of egg yolk, the highest concentration was 0.87 mg/kg.

Some samples of baby food prepared from poultry had relatively high levels of DDE and DDD. These were canned products and contained no DDT because of the conversion of DDT to DDD during the preparation.

Particularly high levels of dieldrin were found in fish from a certain water system. Dieldrin was used in a textile factory for moth protection.

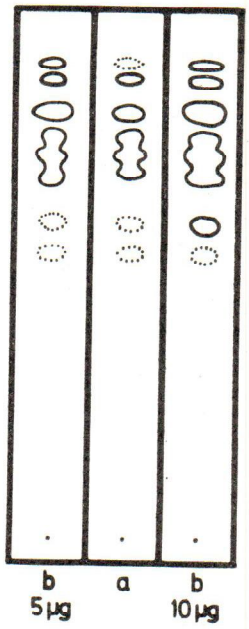

Fig. 5

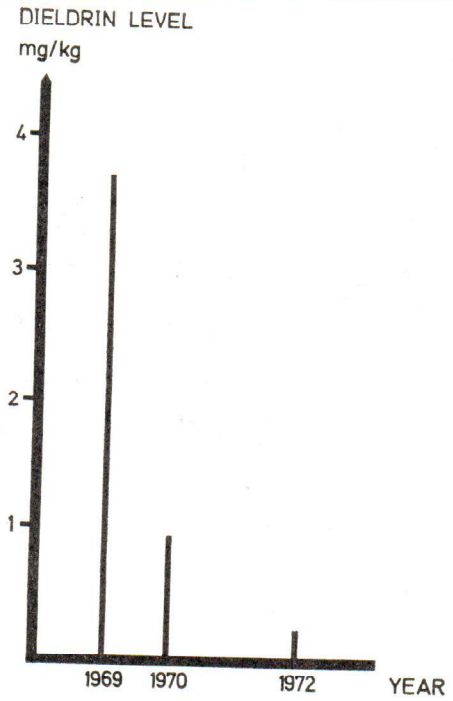

Fig. 6

Fig. 5. *Thin-layer chromatograms of a) PCB from a cardboard box of a breakfast cereal and b) PCB, Clophen A 30*

Fig. 6. *Average concentration of dieldrin in the eel from Klosterfjorden or from the mouth of the Viskan at different times, mg/kg fish flesh*

Fish from the river and a lake downstream from the factory were found to be highly contaminated. Levels as high as 12 mg/kg were found. The sale of fish from this water system was prohibited in 1969.

The factory stopped using dieldrin and there has been a considerable decrease in the levels of dieldrin residues during 1970—1972. In 1969 the average concentration of dieldrin in eel from the river mouth was 3.7 mg/kg, in 1970 0.94 mg/kg, and in 1972 0.23 mg/kg (Fig. 6). The prohibition has now been lifted for some lean fish.

In the investigation of about 1000 samples of fish during the period 1967—1970 (7), lean fish (pike, perch, cod, haddock) showed low levels of pesticides and PCB. Fat fish (eel, herring, salmon) had higher levels of DDT (including the metabolites DDE and DDD), dieldrin, and PCB than other animal products. The fish from the Baltic Sea are especially contaminated. The Baltic herring is a common constituent of the Swedish diet. The herring from certain parts of the southern Baltic Sea contained high levels of DDT and its metabolites (average 3 mg/kg). High levels of

dieldrin and PCB were found in the herring from the Stockholm archipelago (average 0.046 mg dieldrin and 0.9 mg PCB/kg). The herring from the nothern parts of the Baltic (the Bothnina Bay) had much lower levels of pesticides and PCB.

The cod from the Baltic sea contained low levels of pesticides and PCB. Cod is a lean fish (about 0.5% fat), but as it is next to herring in the food chain, it has a large intake of pesticides. The liver of cod from the Baltic contained very high levels of DDT. Concentrations of up to 22 mg kg of DDT+DDE+DDD have been determined. Cod liver from the Baltic was declared unfit for human consumption in 1971.

The use of DDT, aldrin and dieldrin has been prohibited in Sweden since 1st January 1970. To study the possible effects of the restrictions we have analysed human milk from the Mother's Milk Central in Stockholm and found that the DDT level in human milk has decreased by 50% from 1967 to 1972 (8). The average concentration in 1967 was 0.04 mg/kg

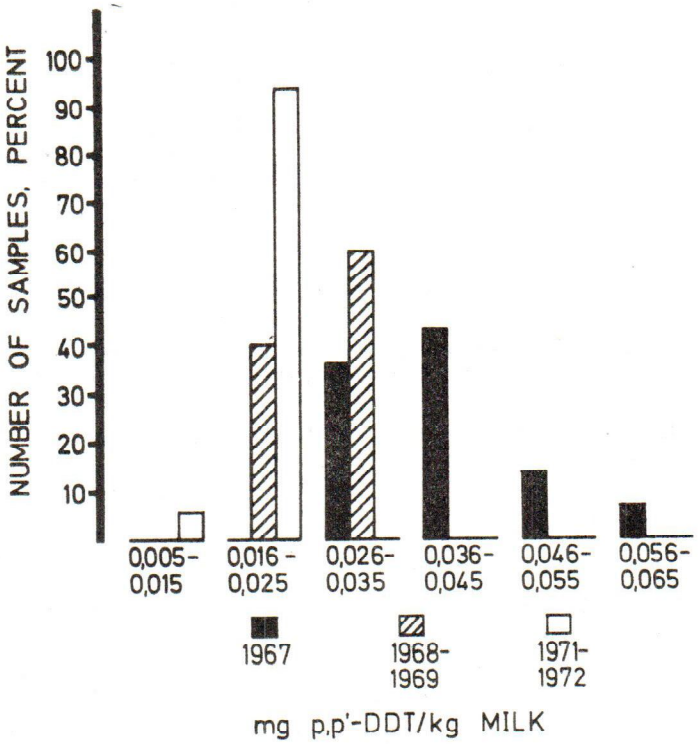

Fig. 7. *Diagram of the distribution at different levels of p,p'-DDT in human milk from the Mother's Milk Central in Stockholm (pooled samples, each from 10—20 mothers) in 1967 (14 samples), 1968—69 (5 samples), and 1971—72 (18 samples). The number of samples having levels within given ranges is expressed in per cent of the total number of samples within the same time interval*

milk (1.3 mg/kg milk fat) and in the latest investigation (November 1971 — January 1972) the concentration was 0.02 mg/kg milk (0.6 mg/kg milk fat). Even before the prohibition a decrease in the DDT level was obvious (Fig. 7). Probably the public discussions at the time about the harmful effects of the substance led to a reduced use of DDT as a household insecticide. The average DDE concentration was unchanged, about 0.06 mg/kg milk. The average levels of other pesticides found in the human milk 1971—1972 were DDD 0.0007, dieldrin 0.0018, α-BHC 0.0002, β-BHC 0.0053, lindane 0.0005, and hexachlorobenzene 0.0059 mg/kg (8).

The average level of polychlorinated biphenyls in human milk seems to have increased (Fig. 8). In 1967 the average PCB level was 0.014 mg/kg milk, in November 1971 — January 1972 the average concentration was 0.025 mg/kg milk (8).

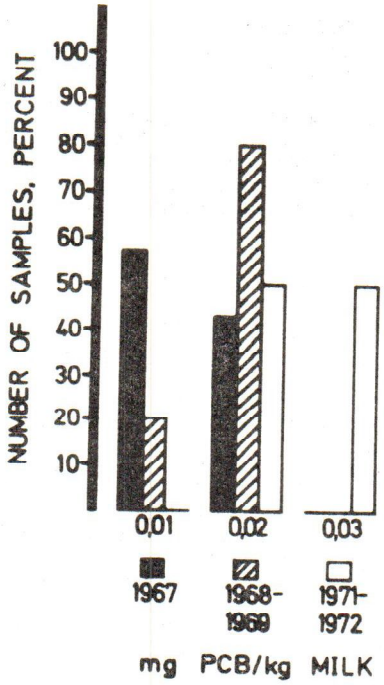

Fig. 8. *Diagram of the distribution at different levels of PCB in human milk from the Mother's Milk Central in Stockholm (pooled samples, each from 10—20 mothers) in 1967 (14 samples), 1968—69 (5 samples), and 1971—72 (18 samples). The number of samples having levels within given ranges is expressed in per cent of the total number of samples within the same time interval*

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Sažetak

PESTICIDI IZ SKUPINE KLORIRANIH UGLJIKOVODIKA I POLIKLORIRANI BIFENILI U HRANI U ŠVEDSKOJ

U ovom radu autor je prikazao rezultate mjerenja sadržaja pesticida iz skupine kloriranih ugljikovodika i polikloriranih bifenila u hrani za razdoblje od 1967. do 1972. godine.

Uzorci povrća sadržavali su samo izuzetno kloriranih ugljikovodika u količinama većim od onih dopuštenih u Švedskoj. Od životinjskih proizvoda jedino je mast riba iz nekih voda sadržavala visoke koncentracije pesticida i polikloriranih bifenila. Najveće koncentracije DDT-ja i njegovih metabolita nađene su u jetri bakalara iz Baltika. Neki uzorci cerealija bili su kontaminirani polikloriranim bifenilima iz materijala za pakiranje.

U ribama iz pojedinih područja očito je smanjena koncentracija dieldrina kao i p,p'-DDT-ja u majčinu mlijeku štokholmskog područja. Povećana je prosječna razina polikloriranih bifenila u majčinom mlijeku.

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