Residues of Organochlorine Insecticides and PCBs in Samples of Norwegian Butter 1966-1972

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Samples of Norwegian butter have been subjected to monthly analyses with regard to organochlorine insecticides in alternate years since 1966 as part of a programme to map the occurrence of this type of contamination in food stuffs in Norway. The ban on DDT which came into effect on 1st October 1970, gave the chance to study the effect of a local stop in the supply of DDT. Results from investigations in the various years have been published elsewhere (SAKSHAUG 1968, BJERK & SAKSHAUG 1969, BJERK 1971, BJERK & KVESETH 1974).

At the onset of the investigations, dairies were chosen which supplied the most populated parts of Norway. After a while interest came to be concentrated on Western Norway where relatively high concentrations of DDT were found. Analyses of cod liver from certain fjords in Western Norway also show fairly high levels of DDT (BJERK 1973, STENERSEN & KVALVÅG 1972). These high values are probably the result of the widespread use of DDT in fruit orchards.

Materials and methods

Two types of butter, dairy butter and farm butter, were analysed. The latter type is produced in small quantities only, often by the individual dairy farmer. Several dairies participated in the investigations, between 4 and 6 each year. Apart from 1972, the samples were taken randomly in order to give an average for the dairy. In 1972, samples were taken from the same area throughout the year, where possible, from the same producer.

The structure of Norwegian dairy farming has changed during the last years. Milk production has become more and more concentrated to specialised units. The risk of contamination with DDT should therefore decrease, as the dairy farmer is no

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longer engaged in other activities, which require the use of DDT. The ongoing rationalisation of the dairy industry into steadily larger units will also reduce the effect an incidental contamination, as butter is produced in larger batches. This is especially true for dairy butter.

Analyses were originally carried out using the socalled DMF method (de FAUBERT-MAUNDER et al. 1964) with minor modifications. In 1970 and 1972, the clean up stage using column chromatography was omitted and instead the extract was divided and the two aliquotients were treated with conc. sulphuric acid and alcoholic potassium hydroxide respectively. The subsequent n-hexane extracts were analysed in gas chromatographs, equipped with EC-detectors. All samples were analysed in 2 columns, one 10 per cent QF 1 and one 4 per cent SF-96 Spiked samples showed a recovery rate of 80-105 per cent for DDT, DDD and DDE at the 0.01-0.05 ppm level.

Results

The annual means for DDT residues found in the investigated dairies were in all cases less than the parctical residue limit established by WHO/FAO for DDT residues in milk fat. At the beginning of the period the annual mean was close to the limit in certain dairies in Western Norway, but the level fell considerably during 4-6 years.

Three dairies participated in all 4 investigations : A (Stavanger), B (Bergen) and C(Trondheim). Results are shown in table 1. The means of positive findings are given. As there were only slight differences between dairy and farm butter from the same dairy, the mean of all the samples is given. Both A and C are situated in agricultural districts, while there is little agricultural activity apart from fruit growing in the district in which B is situated.

Results from other dairies are shown in table II. F is situated in one of the most well known fruit growing areas, and F's butter production comprised about 1.5 per cent of the national total. The high DDT levels made it desirable to investigate levels also in 1972. However, butter production had ceased, and the laboratory received samples from a neighbouring dairy, G. These samples, however, were from a hill farm, and are therefore not representative of the district F-G.

Low levels were found, apart from the November and December samples. These contained 0.8 and 0.5 ppm DDT residues respectively, almost entirely DDT and DDD. This contamination was considered to be incidental. These individual levels raise the mean level for this dairy from 0.020 to 0.15 ppm DDT.

The dairies E, H and I are situated in areas with little agriculture, whilst D is situated in an agricultural district. Results of analyses show a fall in DDT levels from 1968 to 1972, even at these low levels.

TABLE 2

Residues of organochlorine insecticides and PCB's in butter from selected Norwegian dairies, ppm.

Year	L	N	Dieldrin		DDT		DDD		DDE		total DDT		PCB's	
			p	mean	p	mean	p	mean	p	mean	p	mean	p	mean
1968	D	9	9	0.016	8	0.043	8	0.013	9	0.016	9	0.068	0	
"	E	20	20	0.020	19	0.041	15	0.015	18	0.010	20	0.058	0	
1970	F	18	14	0.033	18	0.19	18	0.37	18	0.24	18	0.87	2	0.01
1972	G	11	10	0.009	7	0.096	3	0.22	11	0.016	11	0.15	11	0.039
"	H	18	17	0.016	6	0.004	4	0.006	16	0.006	17	0.008	17	0.040
"	I	24	22	0.011	13	0.007	6	0.005	23	0.006	24	0.012	24	0.034

L: Locality, D: Oslo, E: Tromsø, F: Sogn, G: Vik, H: Bolsøy, I: Sand
N: no. of samples, p: no. of samples with detectable residues.

Average levels for dieldrin remained fairly constant in all analytical series. This was not unexpected as this insecticide has only been used to a very limited extent in Norway. BRO-RASMUSSEN et al. (1968) found 0.03 ppm dieldrin in Danish butter in 1966, and reckoned that between 60 and 100 per cent of this dieldrin was derived from feed and feed concentrates, mainly imported products. Whilst DDT residues in Norwegian butter samples in 1972 constituted about 2 per cent of the practical residue limit recommended by FAO/WHO, dieldrin residue amounted to about 12 per cent of the FAO/WHO limit.

PCBs were demonstrated in samples from 1970 and 1972. Though concentrations have not increased, PCB residues were more common in the 1972 material. FRIES (1972) has reported PCB concentrations of between 1 and 10 ppm in butter fat. The low values and uniform distribution found in the Norwegian material suggests a more indirect source of contamination. An attempt to clarify this will be made during the investigation in 1974.

Discussion

Average values were not calculated in 1966, though these can be estimated from a histogramme of the monthly results. If these estimates are used, it will be found that, as regards localities A and C in 1966, there was a maximum between 1966 and 1968. Results from B, where DDT levels were approximately 10 times higher than for A and C in 1966, do not show such a maximum. The fall in DDT levels which is detectable as early as 1966, is probably due to a better understanding of the possible risks associated with the use of DDT. This has probably led to better storage, use and disposal of this chemical. The ban on the use of DDT from October 1970 caused an acceleration in the fall in DDT levels. Whilst DDT levels fell by about 50 per cent between 1968 and 1970, they fell by 75-96 per cent between 1970 and 1972.

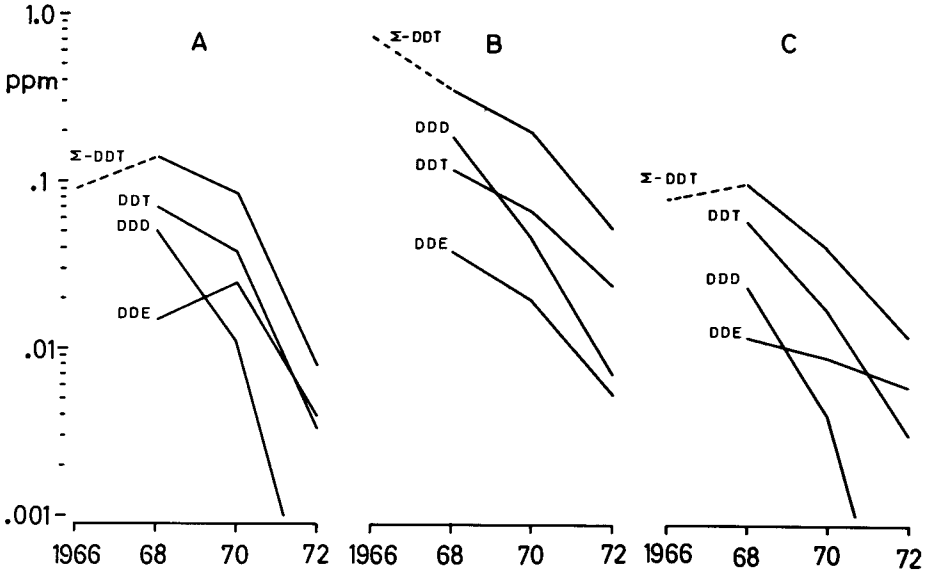

Figure 1. Residues of DDT, DDE and DDD in butter from three Norwegian dairies 1966-1972.

The amount of DDT, DDD and DDE in samples from A, B and C during the years 1968-1972, are given in fig. 1. Mean values are given on the basis of the sum total of samples, and non-detectable concentrations are given as 0. All three localities

show about the same distribution. In 1968, DDD was dominant in samples from B. This would seem to indicate that animal feed contained large amounts of DDT, a fact which was confirmed on analyses of grass from these fruit orchards in 1969, when 1-50 ppm DDT were demonstrated (SØMME et al.1970). When DDT is administered experimentally to cows, only DDD and DDT are found in the milk, DDD levels being higher than DDT levels (FRIES 1969, SMART & ROUGHAN 1972). When supplies of DDT are cut off, while both DDD and DDT concentrations fall, the concentration of DDD falls the faster. This development can be followed at all three localities.

The fall in DDE was quite moderate during the period 1968-1972. The greatest fall was seen in samples from B, though even here it was less than a factor of ten. The present development indicates that mainly DDE residues will be found in Norwegian butter samples in the future, something which can already be ascertained in the samples from A and C in 1972.

DDE must be taken as such if it is to be demonstrated in cow's milk, and the excretion rate is about the same as for DDT (FRIES et al 1968). As the fall in DDE in the material from A to C was less than for DDT, this would seem to indicate a background level. It will be interesting to see if DDE levels in 1974 fall below 0.005 ppm.

The low residue concentrations in the 1972 samples indicate that animal feed stuffs were contaminated only to a very slight degree. STULL et al.(1968) investigated the excretion of DDT residues into the milk in cows fed on a ration containing 0.09-0.65 ppm DDT residues for a period of 17 months. They found 1.2-4.3 ppm DDT in milk fat. WHITING et al.(1972) demonstrated a fall of 90 per cent to approximately 0.25 ppm in milk after 10 months use of feed stuffs containing less than 0.01 ppm DDT residues. LABEN (1968) mentions in a survey, that when only slightly contaminated feed (0.02-0.05 ppm) is used, milk will contain 20-30 times that concentration of DDT, and that absorption and excretion of DDT into the milk is more effective at such low concentrations than is the case with larger doses of DDT.

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