PAPERS AND ORIGINALS

Organochlorine Pesticide Residues in Human Fat in the United Kingdom 1969-71

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Summary

During the period 1969-71 samples of human body fat were taken during routine necropsies on 201 subjects over 5 years old and 20 stillborn babies and infants under 5 years old, and analysis was carried out for organochlorine pesticide residues. By comparing the results from this period with those from similar studies undertaken in 1963-4 and 1965-7, it is clear that a downward trend continues.

Introduction

Studies on the residues of organochlorine pesticides in human body tissues have attracted considerable attention in many countries, especially over the past 10 years. The determination of national mean concentrations of these materials in human fat serves as a convenient and probably valid index of the overall degree of exposure of a population to these compounds. Useful information is available from each set of results, but possibly greater value derives from study of any trends that may become apparent over a period of years. Such studies may usefully show the effect of the current general policy of reducing the use of certain persistent organochlorine pesticides or of limiting their application to those situations where there are no satisfactory substitutes (F.A.O./W.H.O., 1970; Department of Education and Science, 1969). In the past five years many publications have appeared which give the results of analyses of samples of human fat carried out in countries ranging from Australia (Wassermann *et al.*, 1968) and New Zealand (Dacre, 1969) to Denmark (Weihe, 1966), and from Israel (Wassermann

et al., 1967) via Hungary (Berend *et al.*, 1970) and Holland (de Vlieger *et al.*, 1968) to Hawaii (Casarett *et al.*, 1968). All of these papers repeat the same story; residues of several organochlorine pesticides are always present, the only difference lying in the relative mean concentrations found. Comparison between results obtained in different countries shows that for DDT and its derivatives there is wide variation, but there is a general consistency in the levels of BHC and dieldrin detected. Unfortunately in few of these studies were enough samples examined for statistical appraisals to be justified.

The study reported here was undertaken jointly by the Laboratory of the Government Chemist (Department of Trade and Industry) and the Department of Health and Social Security on behalf of the Panel for the Collection of Residues Data of the Scientific Sub-Committee of the Advisory Committee on Pesticides and Other Toxic Chemicals. In its mode of operation it was very similar to the studies carried out in 1963-4 (Egan *et al.*, 1965) and in 1965-7 (Abbott *et al.*, 1968), but on this occasion the field was further extended to include Northern Ireland and also parts of south-west England; samples were also received from an additional centre in Scotland.

Methods and Materials

Between January 1969 and March 1971, 201 samples of human perirenal fat, each of not less than 5 g, were collected at necropsy from subjects over 5 years old during the course of routine examination by pathologists. In addition 20 further necropsy samples were received from either stillborn or prematurely born babies or from infants under 5 years old. All specimens were placed in specially cleaned and prepared glass containers and stored at -20°C while awaiting analysis. The samples were examined as described in the report of the second study (Abbott *et al.*, 1968) except that in most cases the final determination of the residues by gas-liquid chromatography involved the use of at least one additional stationary phase. As well as the silicone and Apiezon L columns regularly used for this purpose, columns of the more polar cyanosilicone GE-XE 60 (Simmons and Tatton, 1967) or of 3% diethylene glycol succinate on silanized Chromosorb P (Thomson *et al.*, 1969) were found to be of value in supplying additional confirmation of the presence and amounts of some residues. On a number of the samples the identities of the observed residues of dieldrin, *pp'*-DDE, and *pp'*-DDT were

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confirmed by thin-layer chromatography followed by the preparation of chemical derivatives (Abbott *et al.*, 1969).

Results

The combined results for all samples from subjects over 5 years old are summarized in Table I. For each residue the distribution among the samples tends to be skew, and geometric means are possibly more meaningful than arithmetic means when comparison with other data is required; these have therefore been included for the major residues. As in other human fat studies, an interesting feature of the results is that beta-BHC was by far the most abundant of the BHC isomers detected. It was also of almost universal occurrence, as were trace amounts of hexachlorobenzene. Identification of the latter was based largely on the agreement found between the results from the various gas-liquid chromatography systems, although for some samples additional confirmation was provided by the behaviour of the compound on column chromatography or thin-layer chromatography. Heptachlor epoxide was identified solely by the evidence of gas-liquid chromatography, the amounts present being too small for ready confirmation by chemical means.

The gas-liquid chromatograms from most samples included a few minor peaks which were suggestive of the presence of traces of polychlorobiphenyls. The compounds responsible for these could be separated from the organochlorine pesticide residues (except hexachlorobenzene and *pp'*-DDE) and estimated in terms of polychlorobiphenyls by a procedure frequently used for wildlife sample extracts (Report of the Government Chemist, 1969; Collins *et al.*, 1972). The amounts present were in general far too small to justify this as a regular feature of the analysis and in no sample exceeded an estimated 1 mg/kg. The presence of these compounds in human tissues was indicated by Widmark (1967) and by Acker and Schulte (1970).

As is common to all studies of organochlorine residues in human fat, *pp'*-DDT together with its breakdown product *pp'*-DDE formed the major part of all residues found. Very

minor amounts of the isomer *op'*-DDT were occasionally seen and *pp'*-TDE was also infrequently seen; overall these other derivatives amounted to less than 2% of the total DDT equivalent—that is, *pp'*-DDT plus *pp'*-DDE calculated as *pp'*-DDT.

The main interest in these human fat studies, however, is not so much in the results themselves as in the evidence which they may present of changes in residue levels with time. In Table II the summarized results for subjects over 5 years old, subdivided according to sex, are shown for all three studies carried out by this laboratory. Comparison of these figures shows that there is a downward trend for all the main residues observed. If the results from the latest study are compared with those obtained in 1965-7 the decrease in the dieldrin content, based on the geometric means for all samples, is equivalent to about 30% of the earlier value (0.17 mg/kg with 95% confidence limits of 0.15 to 0.19 mg/kg), while the decrease for total DDT is slightly over 20% of the earlier value (2.5 mg/kg, with 95% confidence limits of 2.3 to 2.7 mg/kg). The decline in total DDT appears to result from a proportionately substantially greater decrease of *pp'*-DDT than of *pp'*-DDE. Geometric means for these individual compounds were not published with the results of the earlier studies, but a comparison of the available arithmetic means shows a lowering of the proportion of *pp'*-DDT in the total DDT from 35% in 1963-4 to 26% in 1965-7 and to 21% in the present study.

The decrease in the dieldrin content of human fat is in keeping with the general decline observed for this compound in food and for home-produced mutton fat in particular (Report of Government Chemist, 1970), since the voluntary restrictions placed on the use of this pesticide in the United Kingdom in January 1966 (Ministry of Agriculture, Fisheries and Food, 1964). The decrease in DDT content occurred before the later recommendations of the Wilson Committee (Department of Education and Science, 1969) could have been put into effect but may nevertheless be correlated with the decline, reported over the years, in amounts of DDT compounds in some foods, especially butter (Reports of the Government Chemist, 1963-71).

TABLE I—Pesticide Residues in Human Fat in the United Kingdom 1969-71. Summarized Results for 201 Subjects over 5 Years Old (mg/kg)

	Total BHC	Heptachlor Epoxide	Dieldrin (HEOD)	<i>pp'</i> -DDE	<i>pp'</i> -DDT	Total DDT*	Hexachlorobenzene
Arithmetic mean	0.29	0.03	0.16	1.8	0.52	2.5	0.05
Range	0.0-0.88	0.0-14	0.0-68	0.07-7.5	0.2-6	0.08-9.5	0.0-29
Standard error of mean	0.01	—	0.01	0.09	0.02	0.1	—
Median value	0.26	—	0.13	1.6	0.45	2.2	—
Geometric mean	0.24	—	0.12	1.4	0.40	1.9	—
95% Confidence limits for geometric mean	0.22-0.27	—	0.11-0.13	1.2-1.5	0.36-0.46	1.7-2.2	—

*The total DDT has been calculated by adding to the *pp'*-DDT found as such the *pp'*-DDT equivalent of the *pp'*-DDE found.

TABLE II—Distribution of Organochlorine Pesticide Residues in Fat of Subjects Over 5 Years Old during 1963-71 (mg/kg)

	1963-4		1965-7		1969-71	
	Male (43)	Female (23)	Male (157)	Female (91)	Male (116)	Female (85)
BHC:						
Beta-BHC arithmetic mean	—	—	0.28	0.28	0.27	0.26
Total BHC { Arithmetic mean	0.43	0.40	0.31	0.30	0.30	0.28
Geometric mean	0.35	0.33	0.24	0.25	0.24	0.24
Range	T-1.0	T-1.0	0.01-1.9	0.04-0.81	0.0-88	0.03-0.61
Heptachlor epoxide:						
Arithmetic mean	—	—	0.045	0.032	0.027	0.024
Range	—	—	0.0-40	0.0-0.8	0.0-14	0.0-0.8
Dieldrin (HEOD):						
Arithmetic mean	0.27	0.23	0.23	0.18	0.16	0.16
Geometric mean	0.21	0.21	0.18	0.15	0.12	0.12
Range	T-0.9	T-0.5	0.01-1.0	0.0-50	0.0-68	0.10-0.58
DDT:						
<i>pp'</i> -DDE arithmetic mean	2.2	1.5	2.2	1.6	1.9	1.6
<i>pp'</i> -DDT arithmetic mean	1.2	1.0	0.83	0.68	0.54	0.49
Total DDT { Arithmetic mean	3.7	2.7	3.3	2.5	2.7	2.3
Geometric mean	3.0	2.2	2.8	2.2	2.1	1.7
Range	0.4-8.5	0.2-8.0	0.08-13	0.21-8.1	0.08-9.5	0.14-6.7
Hexachlorobenzene:						
Arithmetic mean	—	—	—	—	0.05	0.05
Range	—	—	—	—	0.0-11	0.01-0.29

T = Trace.

Total DDT = *pp'*-DDT + *pp'*-DDE calculated as *pp'*-DDT.

The effect, if any, of geographical location is shown in Table III. For this purpose the United Kingdom was divided into four areas: (a) England south of a line drawn from the Bristol Channel to the Wash, (b) England north of this line together with Wales, (c) Scotland, and (d) Northern Ireland. Differences according to sex in amounts of DDT compounds are greater in samples from southern England, and, indeed, the mean level for males from this area was significantly higher than for males from the other three areas and also higher than for all female subjects; the overall mean level for females did not differ significantly from the general mean for males from northern England, Scotland, or Northern Ireland. No such geographical variation was observed in mean levels of residues of BHC or dieldrin.

The distributions of the results by age, sex, and residue level are given in Table IV, the figures showing the percentage of the total falling in each age group. For the BHC isomers and dieldrin the mean amounts found in female subjects of 60 years old and under were significantly less than those for females over 60 years old and for all male subjects. Similar sex differentiation was noted in the previous study (Abbott *et al.*, 1968).

The results of the additional analyses of samples from babies and infants under 5 years old are given in Table V. Compared with the results from this age group quoted in the previous study (Abbott *et al.*, 1968) the amounts of organochlorine compounds found were very much less, showing a general reduction of about 50% and even more for dieldrin residues.

Conclusions

It is again gratifying to record that the mean concentrations of the residues of the three main organochlorine pesticides found in human fat—namely, BHC, dieldrin, and DDT—have continued to decrease over recent years. It is worth repeating that a similar decline has been concurrently seen in the amounts of these compounds occurring in some fatty foodstuffs consumed in the United Kingdom, thus supporting the previously expressed belief that dietary sources are the main factor in determining the residue levels in human fat in this country (Abbott *et al.*, 1968).

As mentioned above, the problem of organochlorine pesticide residues in human tissues is world wide. The results of the present survey for subjects over 5 years old are compared in Table VI with some of those reported elsewhere in recent years. Results for DDT compounds are very variable. The United Kingdom, however, is clearly at the lower end of such a "league table," and, indeed, there has been a decline since 1963-4 in the general level of such pesticide residues. In all these circumstances the situation in the United Kingdom can be regarded as remaining comparatively satisfactory and still improving.

We should like to thank those pathologists throughout the country who made this work possible by providing suitable samples, and those members of the staff of the Laboratory of the Government Chemist who carried out the analysis of the samples. Thanks are also due to Mr. O. F. Newman for the statistical

TABLE III—Arithmetic Mean of Pesticide Residues in Human Fat by Geographical Location of Subjects 1969-71 (mg/kg)

	Southern England*		Northern England and Wales†		Scotland‡		Northern Ireland§	
	Male	Female	Male	Female	Male	Female	Male	Female
Beta-BHC	0.31	0.27	0.23	0.24	0.26	0.24	0.25	0.36
Total BHC	0.34	0.29	0.26	0.26	0.28	0.25	0.29	0.40
Heptachlor epoxide	0.038	0.029	0.023	0.020	0.014	0.018	0.009	0.010
Dieldrin (HEOD)	0.19	0.17	0.14	0.16	0.18	0.15	0.08	0.06
Total DDT	3.2	2.5	2.2	2.1	2.3	1.9	2.4	2.3
pp'-DDE	2.4	1.8	1.5	1.5	1.6	1.3	1.6	1.4
pp'-DDT	0.58	0.51	0.48	0.51	0.51	0.45	0.60	0.44
Hexachlorobenzene	0.046	0.045	0.047	0.053	0.059	0.062	0.038	0.045
No. of subjects	49	46	38	20	17	17	12	2

*Bishop's Stortford, Bristol, Chichester, Hertford, Ipswich, London, Portsmouth, Taunton.

†Bangor, Barnsley, Bolton, Carlisle.

‡Bridge of Earn, Carlisle, Dundee.

§Belfast.

TABLE IV—Distribution of Pesticide Residues in Human Fat by Age, Concentration and Sex (1969-71). Results are given as Percentages

Pesticide	Residue Range (mg/kg)	Age Group in Years					
		5-40		41-60		Over 60	
		Male	Female	Male	Female	Male	Female
Total BHC	0-0.10	8	0	6	19	10	7
	0.11-0.20	37	50	18	25	17	18
	0.21-0.30	13	14	18	44	34	24
	0.31-0.40	25	22	31	12	24	22
	Over 0.40	17	14	27	0	15	29
Dieldrin (HEOD)	0-0.10	46	72	34	56	37	24
	0.11-0.20	38	14	27	25	39	45
	0.21-0.30	8	7	24	19	14	16
	0.31-0.40	8	7	9	0	5	4
	Over 0.40	0	0	6	0	5	11
Total DDT equivalent	0-2.0	50	64	31	63	37	46
	2.1-3.0	25	29	21	25	32	16
	3.1-4.0	13	0	21	6	19	24
	4.1-5.0	4	7	9	6	5	9
	Over 5.0	8	0	18	0	7	5
No. of subjects		24	14	33	16	59	55

TABLE V—Organochlorine Pesticide Residues in Fat of Infants under 5 Years Old 1969-71 (mg/kg)

Age Group	No. of Samples		Beta-BHC	Total BHC	Dieldrin (HEOD)	pp'-DDE	pp'-DDT	Total DDT
Prematurely born, stillborn, or died within 24 hours ..	3	Arithmetic mean	0.06	0.07	0.01	0.35	0.06	0.45
		Range	0.05-0.06	0.05-0.10	0.02	0.07-0.62	0.0-0.10	0.08-0.79
1 day to 3 months	8	Arithmetic mean	0.08	0.11	0.03	0.27	0.04	0.35
		Range	0.02-0.17	0.02-0.21	0.07	0.02-0.74	0.0-0.15	0.02-0.98
3 months to 4 years	9	Arithmetic mean	0.08	0.10	0.05	0.32	0.14	0.52
		Range	0.02-0.15	0.03-0.17	0.02-0.10	0.12-0.72	0.02-0.30	0.15-1.04

TABLE VI—Residues in Human Fat in Various Countries

	Period	No. of Samples	Mean Residues (mg/kg)				References
			Total BHC	Dieldrin (HEOD)	pp'-DDE	pp'-DDT	
U.K.	1963-4	65	0.42	0.26	2.0	1.1	Egan <i>et al.</i> (1965)
New Zealand	1963-4	45	0.50	—	—	—	Dacre (1969)
Denmark	1965	18	—	0.20	2.5	0.60	Weihe (1966)
Italy	1965	9	—	—	2.86	1.46	Kanitz and Castello (1966)
Australia	1965-6	12	0.68	0.67	6.3	2.5	Wassermann <i>et al.</i> (1968)
Israel	1965-6	133	—	—	9.9	8.2	Wassermann <i>et al.</i> (1967)
U.K.	1965-7	248	0.31	0.21	2.0	0.78	Abbott <i>et al.</i> (1968)
California, U.S.	1967	64	—	—	16.1	4.8	Rappolt and Hale (1968)
Florida, U.S.	1967	42	—	0.21	6.69	2.77	Radomski <i>et al.</i> (1968)
Hawaii, U.S.	1967-8	11	—	0.03	4.64	1.33	Casarett <i>et al.</i> (1968)
Holland	1967-8	11	—	0.17	1.70	0.32	de Vlieger <i>et al.</i> (1968)
Canada	1967-8	51	—	0.12	4.16	1.56	Kadis <i>et al.</i> (1970)
Arizona, U.S.	1967-8	70	—	0.14	4.58	1.54	Morgan and Roan (1970)
Florida, U.S.	1968-9	90	—	—	5.5	2.3	Davies <i>et al.</i> (1969)
Hungary	1969	—	2.3	—	6.74	5.57	Berend <i>et al.</i> (1970)
Federal Republic of Germany	1970	20	0.45	—	2.2	1.1	Acker and Schulte (1970)
U.K.	1969-71	201	0.29	0.16	1.8	0.52	Present paper

analyses of the results. This report is made by permission of the Government Chemist (Department of Trade and Industry) and the Department of Health and Social Security.

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Varicose Veins, Deep Vein Thrombosis, and Haemorrhoids: Epidemiology and Suggested Aetiology

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Summary

Current concepts on the aetiology of varicose veins, deep vein thrombosis, and haemorrhoids have been examined and, in the light of epidemiological evidence, found wanting.

It is suggested that the fundamental cause of these disorders is faecal arrest which is the result of a low-residue diet.

Introduction

Varicose veins and haemorrhoids are among the most common ailments of economically developed countries. Varicose veins are present in 10-17% of adults in England (de Takatas and

Quint, 1930; Dodd and Cockett, 1956; Martin *et al.*, 1956; Allen *et al.*, 1962) and in North America (Davis, 1968). The Ministry of Pensions and National Insurance (1962) recorded that just under four million working days were lost in Britain in 1959-60 due to these two conditions.

The incidence of deep venous thrombosis in hospital patients varies with the criteria used in diagnosis. Lambie *et al.* (1970), using ¹²⁵I-labelled fibrinogen, showed that nearly half of those they termed "high risk" surgical patients in Britain developed some degree of venous thrombosis. Kakkar *et al.* (1970) estimated its incidence as 20-30% of all surgical patients and 40-45% in older patients undergoing major operations. Deep vein thrombosis is, moreover, no less common in severe medical illnesses (Nicolaidis *et al.*, 1971). Flanc *et al.* (1969) found that about 35% of patients over the age of 40 developed thrombosis in spite of conventional prophylactic measures. Besides being an important cause of postoperative disability, venous thrombosis is the root cause of one of the most dreaded disasters of surgery—pulmonary embolism, which kills about 2,500 people in Britain annually (General Register Office, 1969). Some degree of pulmonary embolism is believed to occur in half of all patients who develop ileofemoral thrombosis (Mavor, 1969). More-

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