CLINICAL RESEARCH

Organochlorine pesticide residues in human fat in the United Kingdom 1976-7

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Abstract

Between May 1976 and August 1977 samples of human body fat were taken during routine necropsies in the United Kingdom on 236 subjects aged over 5 years and four infants aged under 4 months. Comparison with results from earlier studies showed a further decline in residues of *pp'*-dichlorodiphenyltrichloroethane (*pp'*-DDT) and dieldrin (HEOD) and increased amounts of hexachlorobenzene residues; concentrations of other compounds were similar to those observed in the studies carried out in 1963-4, 1965-7, and 1969-71. Comparison of the data with those from other countries showed that the concentrations of organochlorine pesticide residues and polychlorobiphenyls in human fat samples from residents of the United Kingdom remain among the lowest in Europe and, indeed, the world.

Introduction

Studies of the occurrence of trace amounts of organochlorine pesticides and similar compounds in human body tissues continue to attract attention in many countries. The national mean concentrations of these substances have been taken as offering a convenient and valid index of the average overall extent of exposure of the population. Each set of results is of value in itself, but repeated studies at intervals of some years allow trends in concentrations to be observed, which may be correlated with known or suspected changes in usage and availability of the

compounds concerned. The study reported here is the fourth in a series designed and carried out in similar ways, the earlier work¹⁻³ covering 1963-4, 1965-7, and 1969-71.

Experimental methods and materials

Between May 1976 and August 1977, 236 samples of human body fat were collected at necropsy from subjects over 5 years old during the course of routine examinations by pathologists throughout the United Kingdom. In addition, four similar samples from infants less than 4 months old were received. All specimens were placed in specially cleaned and prepared glass phials and stored at -20°C until analysis. The samples were examined by the gas-liquid chromatographic procedure recommended for analysis of animal fats by the Committee for Analytical Methods of the Ministry of Agriculture, Fisheries, and Food.⁴ All extracts after clean-up were fractionally eluted from silica gel to separate polychlorobiphenyls, hexachlorobenzene, and most of the *pp'*-DDE (a derivative of dichlorodiphenyltrichloroethane (DDT)) from the other residues.⁵ The semi-quantitative assessment of the concentration of polychlorobiphenyls was based on a comparison with Aroclor 1260.⁶ The identities of the major residues found were confirmed in some selected extracts by a range of chemical and physical means.⁶

Results and discussion

Table I shows the combined results for all samples obtained from subjects over 5 years old. For each residue the distribution of results among the samples was usually skew, and geometric means are possibly more meaningful than arithmetic means when comparison with other data is required; these were therefore included for the major residues.

Table II shows the results for the subjects over 5 years old subdivided according to sex, together with those from the earlier studies in this series.¹⁻³ The only residue for which a pronounced reduction was apparent was *pp'*-DDT, and this probably reflects the restrictions on the use of DDT imposed in many countries, including the United Kingdom, over the past few years. The gradual reduction in total DDT since 1963 is almost entirely due to this decrease in *pp'*-DDT, the concentration of *pp'*-DDE having remained fairly constant since the studies began. Perhaps not surprisingly, residual concentrations of *pp'*-DDE have not yet been influenced by the decreased use of DDT, as its rate of elimination is low.⁷ A parallel decrease has been

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TABLE I—Residues of organochlorine compounds in human fat (mg/kg) in the United Kingdom, 1976–7. (Results obtained in 236 subjects over 5 years old)

	Beta-HCH	Total HCH	Heptachlor epoxide	Dieldrin (HEOD)	pp'-DDE	pp'-DDT	Total DDT *	Hexachlorobenzene	PCB
Arithmetic mean	0.31	0.33	0.03	0.11	2.1	0.21	2.6	0.19	0.7
Range†	T-1.2	T-1.2	T-0.12	T-0.49	0.03–15	T-2.4	0.04–17	0.02–3.2	T-10
Standard error of mean	0.01	0.01	0.01	0.01	0.12	0.01	0.15	0.01	0.05
Median value	0.29	0.31	0.09	0.09	1.7	0.17	2.1	0.15	0.7
Geometric mean	0.24	0.27	0.09	0.09	1.5	0.15	1.9	0.15	0.6
95% confidence limits for geometric mean	0.22–0.27	0.24–0.29		0.08–0.10	1.3–1.7	0.14–0.17	1.6–2.1	0.14–0.17	0.5–0.6

*Total DDT was calculated by adding to the pp'-DDT found as such the pp'-DDT equivalent of the pp'-DDE and pp'-TDE.

†T is less than 0.01 (less than 0.1 for PCB).

HCH = Hexachlorocyclohexane. DDE = Derivative of DDT. PCB = Polychlorobiphenyls.

TABLE II—Distribution of residues of organochlorine compounds in fat of subjects over 5 years old during 1963–77 (mg/kg)

		1963–4		1965–7		1969–71		1976–7	
		Male (n = 43)	Female (n = 23)	Male (n = 157)	Female (n = 91)	Male (n = 116)	Female (n = 85)	Male (n = 155)	Female (n = 81)
HCH:									
Beta-HCH	Arithmetic mean	..	..	0.28	0.28	0.27	0.26	0.30	0.33
Total HCH	Arithmetic mean	..	..	0.31	0.30	0.30	0.28	0.33	0.35
	Geometric mean	..	..	0.24	0.25	0.24	0.24	0.26	0.29
	Range*	..	..	T-1.0	T-1.0	0.01–1.9	0.04–0.81	0.01–1.2	T-1.1
Heptachlor epoxide	Arithmetic mean	..	..	0.045	0.032	0.027	0.024	0.03	0.03
	Range*	..	..	T-0.40	T-0.08	T-0.14	T-0.08	T-0.09	T-0.12
Dieldrin (HEOD)	Arithmetic mean	..	..	0.23	0.18	0.16	0.16	0.11	0.11
	Geometric mean	..	..	0.21	0.18	0.12	0.12	0.09	0.08
	Range*	..	..	T-0.9	T-0.50	T-0.68	0.10–0.58	T-0.46	0.01–0.49
DDT:									
pp'-DDE	Arithmetic mean	..	..	2.2	1.6	1.9	1.6	2.1	2.1
pp'-DDT	Arithmetic mean	..	..	1.2	1.0	0.83	0.49	0.20	0.23
Total DDT	Arithmetic mean	..	..	3.7	2.7	2.7	2.3	2.6	2.6
	Geometric mean	..	..	3.0	2.2	2.1	1.7	2.0	1.6
	Range	..	..	0.4–8.5	0.2–8.0	0.08–9.5	0.14–6.7	0.13–12	0.04–17
Hexachlorobenzene	Arithmetic mean	..	..	0.05	0.05	0.05	0.05	0.18	0.22
	Range*	..	..			T-0.11	0.01–0.29	0.02–0.68	0.02–3.2
PCB	Arithmetic mean	..	..			†	†	0.8	0.6
	Range*	..	..			All less than 1.0		0.1–10	T-1.5

*T is less than 0.01 (less than 0.1 for PCB).

†PCB was not measured in all cases.

observed since 1966 in the ratio of pp'-DDT to total DDT in the average diet in the United Kingdom.⁹

The slight decrease in dieldrin concentrations probably represents a real effect, as a slow but continuous downward trend is apparent in all studies; this again is probably associated with the successive withdrawals of clearance for the various uses of this pesticide and of aldrin, of which HEOD (dieldrin) is a metabolite. The concentration of hexachlorobenzene found in 1976–7 was higher than that found in the previous study.³ Although this is partly a consequence of better analytical recovery of this compound, this factor alone would account for not more than 30% of the observed increase. The source of these residues is not attributable with any certainty. Hexachlorobenzene is not cleared for use as an agricultural chemical in the United Kingdom. It occurs as an impurity in several substances, however, and is among the waste byproducts of industrial chlorination processes.

Concentrations of polychlorobiphenyls also showed a slight overall increase. Although in the previous study polychlorobiphenyls were measured quantitatively only in those few samples showing the highest concentrations, it was concluded that, in those samples, the concentrations did not exceed an estimated 1 mg/kg. In the present study estimates made on the same basis, though showing a mean value below

TABLE III—Arithmetic means of residues of organochlorine compounds in human fat by geographical location of subjects 1976–7 (mg/kg)

		Southern England*		Northern England and Wales†		Scotland‡		Northern Ireland§	
		Male (n = 74)	Female (n = 43)	Male (n = 51)	Female (n = 26)	Male (n = 22)	Female (n = 9)	Male (n = 8)	Female (n = 3)
Beta-HCH	..	0.29	0.34	0.33	0.33	0.21	0.25	0.49	0.31
Total HCH	..	0.31	0.37	0.36	0.36	0.23	0.27	0.52	0.33
Dieldrin (HEOD)	..	0.11	0.11	0.13	0.13	0.09	0.08	0.10	0.08
pp'-DDE	..	2.3	2.7	2.0	1.5	1.7	1.6	2.9	1.2
pp'-DDT	..	0.20	0.27	0.21	0.21	0.16	0.15	0.29	0.11
Total DDT	..	2.8	3.3	2.5	1.9	2.1	2.0	3.6	1.5
Hexachlorobenzene	..	0.18	0.27	0.17	0.17	0.13	0.17	0.26	0.19
PCB	..	0.7	0.7	0.7	0.5	0.6	0.6	2.0	0.4

*Aylesbury, Bishop's Stortford, East Grinstead, London, Reading, St Austell, Salisbury, Sevenoaks, Winchester.

†Leeds, Walsall, Warwick, Wolverhampton, Bangor, Cardiff.

‡Aberdeen, Dundee, Edinburgh.

§Belfast.

TABLE IV—Distribution of residues of organochlorine compounds in human fat by age, concentration, and sex (1976–7). Results are given as percentages

Residue range (mg/kg)	Age group (years)					
	5–40		41–60		Over 60	
	Male (n = 26)	Female (n = 8)	Male (n = 49)	Female (n = 15)	Male (n = 80)	Female (n = 58)
Total HCH						
0–0.10	35	12	12	20	1	12
0.11–0.20	23	25	12	27	25	14
0.21–0.30	15	37	27	20	11	16
0.31–0.40	8	13	31	26	15	19
Over 0.40	19	13	18	7	48	39
Dieldrin (HEOD)						
0–0.10	73	87	47	66	44	52
0.11–0.20	19	13	47	27	41	33
0.21–0.30	8		2	7	11	7
0.31–0.40			4		3	5
Over 0.40					1	3
pp'-DDE						
0–1.0	46	63	15	40	14	39
1.1–2.0	27	25	51	14	25	21
2.1–3.0	23		10	20	31	12
3.1–4.0	4	12	10	13	19	12
4.1–5.0			10		5	9
Over 5.0			4	13	6	7
pp'-DDT						
0–0.20	81	75	61	67	44	57
0.21–0.40	19	25	35	13	41	30
0.41–0.60			2	13	14	5
0.61–0.80			2	7		3
0.81–1.0					1	3
Over 1.0						2
Total DDT						
0–1.0	38	50	15	20	13	35
1.1–2.0	31	25	39	34	19	17
2.1–3.0	15	13	16	7	26	17
3.1–4.0	12		12	13	19	7
4.1–5.0	4	12	8	13	12	10
Over 5.0			10	13	11	14
Hexachlorobenzene						
0–0.10	38	37	23	7	19	22
0.11–0.20	35	37	51	47	40	35
0.21–0.30	23	13	20	33	21	26
0.31–0.40	4	13	2	13	15	12
Over 0.40			4		5	5
PCB						
0–0.2	4	38	4		3	10
0.3–0.4	23	13	14	20	11	15
0.5–0.6	19	13	17	20	15	28
0.7–0.8	35	12	20	27	25	9
0.9–1.0	11	12	25	13	21	14
Over 1.0	8	12	20	20	25	24

1 mg/kg, exceeded this figure in about 16% of the samples and, apart from the unusually high value of 10 mg/kg found in one sample, reached a maximum of 1.6 mg/kg. Similar increases have been observed in Norway¹⁰ and Finland,¹¹ while a possible slow increase in concentrations of polychlorobiphenyls in human milk has been reported in Sweden.¹² For these compounds, as with *pp'*-DDE, elimination from fat deposits is probably slow, so that it may take some years before the reduced scale of manufacture and use is reflected in a downward turn in concentrations in human fat.

Table III shows the association of concentrations with geographical location. For this purpose the United Kingdom was divided into four areas: England south of a line drawn from the Bristol Channel to the Wash; England north of this line together with Wales; Scotland; and Northern Ireland. Differences according to sex in concentrations of beta-hexachlorocyclohexane (beta-HCH), total HCH, *pp'*-DDE, and total DDT were greatest in samples from Northern Ireland, where the mean concentration in male subjects was higher than that in male subjects from the other areas, although this was not so for the female subjects. Far fewer samples were obtained from Northern Ireland than

pp'-DDE and HCH isomers have remained similar to those reported in the 1969-71 survey. Residue concentrations of hexachlorobenzene are now somewhat higher than those previously reported.

Results of similar surveys carried out in member States of the European Community up to 1972 were reviewed by Gatti,¹⁵ while a worldwide list summarising results published up to 1974 was compiled by Wassermann *et al.*¹⁶ Table VI compares results obtained from the present survey with those recently reported elsewhere. These comparisons show that the amounts of organochlorine pesticide residues and polychlorobiphenyls in human fat samples from residents of the United Kingdom remain among the lowest in Europe and, indeed, the world.

We should like to thank the pathologists throughout the country who made this work possible by providing suitable samples, and the members of staff of the Laboratory of the Government Chemist who

TABLE V—Residues of organochlorine compounds in fat of four infants under 4 months old, 1976-7 (mg/kg)

Age	Beta-HCH	Total HCH	Dieldrin (HEOD)	<i>pp'</i> -DDE	<i>pp'</i> -DDT	Total DDT	Hexachlorobenzene	PCB
Newborn	0.07	0.10	0.03	0.54	0.04	0.68	0.04	0.1
2 months	0.10	0.11	0.02	0.50	0.01	0.60	0.05	0.2
3 months	0.03	0.03		0.05		0.06		
3 months	0.15	0.17	0.09	0.92	0.14	1.2	0.13	0.5

TABLE VI—Residues of organochlorine compounds in human fat in various countries

Country	Period	No of samples	Mean residues (mg/kg)						Reference
			Total HCH	Dieldrin (HEOD)	<i>pp'</i> -DDE	<i>pp'</i> -DDT	Hexachlorobenzene	PCB	
Belgium	1975	60	0.76*	0.26	6.5	1.52	1.36		17
Canada	1972	168	0.065†	0.069	2.095	0.439	0.062	0.907	18
Denmark	1972-3	78		0.12	3.7	0.6†		3.8	19
Federal Republic of Germany		282	0.99†‡	0.14†	4.4	1.1	5.6	8.3	20
Greece		50	0.98§	0.23	7.86	1.99	3.84		21
Japan	1974	30	2.36		2.91	0.68		1.04	22
New Zealand .. .	1973	51	0.49†	0.21	5.64	0.70	0.31	0.82	23
New Zealand .. .	1973-4	21			4.4	0.46		0.35	24
Northern Ireland ..	1975	11	0.45	0.10	1.60	0.34	0.15		13
Spain	1977	40	0.062†	0.150	2.268	1.781			25
Switzerland .. .	1971-2	12	0.90*	0.29	3.8	1.6	1.9	1.0	26
USA¶	1973-4	898	0.21*	0.15	4.1†				27
UK	1976-7	236	0.33	0.11	2.1	0.21	0.19	0.7	Present paper

*Present as beta-HCH. †Calculated from authors' data. ‡Based on 50 samples. §Present as lindane (gamma-HCH). ||Based on 35 samples. ¶Expressed as geometric means.

from the other areas, however, although these findings are in general agreement with the observations of Harper *et al.*¹³ regarding concentrations of total HCH. It has been suggested that this difference between Northern Ireland and other areas is due to a higher dietary intake of HCH, mainly from dairy products, as higher concentrations of HCH isomers have been observed¹⁴ in dairy products in Northern Ireland compared with the rest of the United Kingdom. The mean concentrations of beta-HCH and total HCH in samples from Scotland were lower than those in samples from the other areas.

Table IV gives the distributions of the results for each compound by age, sex, and concentration. Fewer results fell in the higher ranges in the subjects aged 5-40 years than in the older subjects; but there was little difference between those aged 41-60 years and those aged over 60 years. The distribution of concentrations of polychlorobiphenyls was more even than the distributions of pesticide concentrations, almost all of which were skew with most results in the lower ranges. The exception was the total HCH residue in the group aged over 60.

Table V gives the results of the analyses of samples from infants under 4 months old, but these were too few to support any realistic conclusions. They do, however, serve to confirm earlier findings that such residues occur in newly born infants.

Conclusions

The previously observed decline in residue concentrations of *pp'*-DDT and dieldrin has continued, while concentrations of

carried out the analysis of the samples. Thanks are also due to Miss Joan Vernon for the statistical analyses of the results. This report is made by permission of the Government Chemist (Department of Industry), the Department of Health and Social Security, and the Ministry of Agriculture, Fisheries, and Food.

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OF INOCULATION.

THOUGH no disease, after it is formed, baffles the powers of medicine more effectually than the small-pox, yet more may be done beforehand to render this disease favourable than any one we know, as almost all the danger from it may be prevented by inoculation. This salutary invention has been known in Europe above half a century; but, like most other useful discoveries, it has, till of late, made but slow progress. It must however be acknowledged, to the honour of this country, that inoculation has met with a more favourable reception here, than among any of our neighbours. It is still however far from being general, which we have reason to fear will be the case, as long as the practice continues in the hands of the Faculty.

No discovery can be of general utility, while the practice of it is kept in the hands of a few. Had the inoculation of the small-pox been introduced as a fashion, and not as a medical discovery, or had it been practised by the same kind of operators here, as it is in those countries from whence we learned it, it had long ago been universal. The fears, jealousies, the prejudices, and the opposite interests of the Faculty, are, and ever will be, the most effectual obstacles to the progress of any salutary discovery. Hence, it is that the practice of inoculation never became, in any measure, general, even in England, till taken up by men not bred to physic. These have not only rendered the practice more extensive, but likewise more safe, and, by acting under less restraint than the regular practitioners, have taught them that the patient's greatest danger arose, not from the want of care, but from the excess of it.

MEDICINE.—The measles being an inflammatory disease, without any critical discharge of matter, as in the small-pox, bleeding is commonly necessary, especially when the fever runs high, with difficulty of breathing, and great oppression of the breast. But if the disease be of a mild kind, bleeding may be omitted.

BATHING the feet and legs frequently in luke-warm water both tends to abate the violence of the fever, and to promote the eruption.

THE patient is often greatly relieved by vomiting. When there is a tendency this way, it ought to be promoted by drinking lukewarm water, or weak camomile-tea.

WHEN the cough is very troublesome, with dryness of the throat, and difficulty of breathing, the patient may hold his head over the steam of warm water, and draw the steam into his lungs.

HE may likewise lick a little sperma ceti and fugar-candy pounded

together; or take now and then a spoonful of the oil of sweet almonds, with fugar-candy dissolved in it. These will soften the throat, and relieve the tickling cough.

IF at the turn of the disease the fever assumes new vigour, and there appears great danger of suffocation, the patient must be bled according to his strength, and blistering-plasters applied, with a view to prevent the load from being thrown on the lungs, where if an inflammation should fix itself, the patient's life will be in imminent danger.

OF THE ERYSIPELAS, OR ST. ANTHONY'S FIRE.

THIS disease, which in some parts of Britain is called *the rose*, attacks persons at any period of life, but is most common between the age of thirty and forty. Persons of a sanguine or plethoric habit are most liable to it. It often attacks young people, and pregnant women; and such as have once been afflicted with it are very liable to have it again. Sometimes it is a primary disease, and at other times only a symptom of some other malady. Every part of the body is liable to be attacked by an erysipelas, but it most frequently seizes the legs or face, especially the latter. It is most common in autumn, or when hot weather is succeeded by cold and wet.

SYMPTOMS.—The erysipelas attacks with shivering, thirst, loss of strength, pain in the head and back, heat, restlessness, and a quick pulse; to which may be added vomiting, and sometimes a delirium. On the second, third, or fourth day, the part swells, becomes red, and small pustules appear; at which time the fever generally abates.

WHEN the erysipelas seizes the foot, the parts contiguous swell, the skin shines; and, if the pain be violent, it will ascend to the leg, and will not bear to be touched.

WHEN it attacks the face, it swells, appears red, and the skin is covered with small pustules filled with clear water. One or both eyes are generally closed with the swelling; and there is a difficulty of breathing. If the mouth and nostrils be very dry, and the patient drowsy, there is reason to suspect an inflammation of the brain.

IF the erysipelas affects the breast, it swells and becomes exceedingly hard, with great pain, and is apt to suppurate. There is a violent pain in the arm-pit on the side affected, where an abscess is often formed.

(Buchan's *Domestic Medicine*, 1786.)