A Survey of Organochlorine Insecticide Residues in Sri Lanka

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INTRODUCTION

It is well known that loss of crop yields due to insect damage is substantial and that the most direct method of insect control is by the use of insecticides. Insecticides are chemicals which are poisonous to insects. They are also poisonous to other forms of life but they may either be much more lethal to insects than to mammals (including man) or it may be possible to apply them in such a way that insects receive lethal doses while other forms of life escape.

Many different chemicals are used to control insects. In general, these chemicals fall into three groups namely organochlorines, organophosphates and carbamates. The organochlorines are generally very persistent while most of the organophosphates persist at most for a few weeks in soil. All available evidence shows that organophosphates are relatively short-lived in biological systems. There is very little data concerning the persistence of carbamates in the environment. According to Metcalf (1966) carbamates are not persistent in biological systems and are readily metabolized and excreted. Several organophosphates and some carbamates however, have higher mammalian toxicities than the organochlorines and are more hazardous to the user. Generally, organochlorine insecticide residues are stored in the tissues particularly in fat (being fat soluble) so that toxicity could result due to a cumulative effect. The persistence of some of these insecticides is mainly due to the fact that they are transformed in the soil or living tissues to equally or even more toxic chemicals. For example, heptachlor turns to heptachlor epoxide a stable substance considerably more poisonous than heptachlor itself; aldrin is transformed into dieldrin which is toxic and stored in the fat by living animals. Although some of the DDT is changed into the much less poisonous DDE and some of it (DDT) is excreted, there is substantial accumulation of DDT in the body. The potential hazards of persistent insecticides to the environment have been dealt in detail by many writers (Carson, 1963; Mellanby, 1970; Edwards, 1976).

In Sri Lanka prior to 1976 organochlorine insecticides such as DDT, BHC, heptachlor, endrin, dieldrin and aldrin were used for agricultural purposes. Of these DDT was also extensively used for the control of malarial mosquitoes since 1946. This was, however, replaced by malathion as the malarial mosquitoes developed resistance to DDT. Fortunately, most organochlorine insecticides have now been replaced by the less persistent organophosphates and carbamates, but BHC and, to a lesser extent, aldrin and chlordane, are still available for agricultural use. Since large amounts of organochlorines have been used in the past a survey was undertaken to determine their residue levels in the Sri Lankan environment and the results are presented in this paper.

MATERIALS AND METHODS

1. *Collection of Samples*

Market sampling of fruits and vegetables such as cabbage, carrots, beetroots was initiated in 1974. Around 300 samples were collected from different parts of Sri Lanka (Table 1). Jaffna was selected to carry out a more detailed survey since farmers here use intensive amounts of pesticides in their cultivation. In this latter survey samples were collected directly from the farmers. Other than fruits and vegetables, samples of soil and water were also obtained.

Processed foods (fruit juices, jams, etc.) submitted by the Marketing Department, tobacco from Ceylon Tobacco Company Limited and spices and coffee from Bureau of Ceylon Standards were also included in the survey.

A few samples of human fat tissues, cow's milk and rice were also analysed.

2. *Procedures*

(i) *Fruits, Fruit juices and Vegetables*

(*a*) Sample preparation

Sample preparation consisted of taking a representative sample, weighing it and if the sample was solid, homogenizing it.

(*b*) Extraction procedure

The acetonitrile procedure of Mills, Onley and Gaither (1963) was followed for the extraction. The extract in acetonitrile was transferred to a separatory funnel and petroleum ether (50–60°C) added. After vigorous shaking and separation, the petroleum ether phase containing the pesticides was dried with anhydrous $Na_2\ SO_4$. This was thereafter concentrated to a small volume in a Büchi rotary evaporator.

(*c*) Clean up procedure

The concentrated extract was sent through a florisil column and different pesticides eluted with 6% and 20% ethyl ether in petroleum ether. Both eluates were reduced to a small volume (10 ml) for injection into the gas chromatograph.

(ii) *Water and Soil*

The samples were extracted with hexane and no clean up procedure was carried out.

(iii) *Milk*

Fat in the milk was extracted using a mixture of methanol, ethyl ether and petroleum ether. This was concentrated to a small volume. Thereafter extraction and clean up carried out as for vegetables (Mills, 1961).

(iv) *Rice*

The extraction was carried out as described in i (b) but using a different ratio of acetonitrile to water. Clean up was done as for vegetables.

3. *Analysis*

The samples were analysed on a Carlo Erba gas chromatograph Model GI Series 450 using an electron capture detector (ECD–HT 20–63 Ni) and coiled glass column (2.5 mx6 mm) packed with a mixture of 1.15% SP 2250 and 1.95% SP 2401. The column temperature was 220 °C while the nitrogen carrier gas flow was 25 ml/min.

RESULTS AND DISCUSSION

In this survey the residues of DDT, lindane, aldrin, dieldrin, endrin, heptachlor and endosulfan were determined. Although both aldrin and dieldrin were detected, they were added together and reported as dieldrin. Very little dieldrin was used in the past while aldrin is still in use to a limited extent. It is therefore likely that the dieldrin could have mostly arisen from the transformation of aldrin to dieldrin.

Of the organochlorine insecticide residues determined, two are of particular interest in terms of their past and present usage in Sri Lanka. The first, DDT was used in agriculture until it was banned in 1972 but its use for malarial vector control continued until 1975. An idea of the quantities used could be obtained from the 1971 import figures according to which some 2.5 million pounds of DDT was used for vector control and about 10,000 pounds of DDT for agriculture. The second organochlorine insecticide BHC is still being used for stem borer and paddy bug control. In this case some 25 tons of total isomers are annually imported.

The all island survey showed that DDT and BHC residues were detected in 60.9 and 42.5% of the vegetable samples respectively. The percentages for dieldrin, endrin, heptachlor and endosulfan were 27.9, 6.1, 13.3 and 22.8 respectively. In the detailed survey carried out in the Jaffna district more samples contained BHC (75.9%) than DDT (53.4%). None of the samples contained endrin while dieldrin, heptachlor and endosulfan were detected in 39.6%, 27.6% and 24.1% of the samples respectively. The levels of these insecticides were low and well below the FAO/WHO recommended residue tolerances (Tables 2, 3 & 8).

Most samples of rice grain obtained from Bombuwela, Bathalagoda and Maha Illuppallama contained lindane and DDT. These values are much lower than the FAO/WHO recommended residue tolerances (Tables 4 & 8). In additon, samples from Bombuwela contained heptachlor while those from Maha Illuppallama had dieldrin. The levels of these two insecticide residues (especially heptachlor) in the grain samples were much higher than the FAO/WHO limits (Tables 4 & 8). It is very likely that these two insecticides must have come from the adjacent uplands where they would have been applied for the control of insects such as termites. The presence of lindane in rice grain is understandable because BHC dust is widely used for the control of paddy bug.

A batch of processed fruits obtained from the Marketing Department indicated the presence of insecticide residues only in the case of pineapple (Table 5). Export products such as spices, tobacco, coffee and tea also contain these insecticide residues but as for vegetables the levels were low (Table 6).

A few soil samples taken from Jaffna contained very low levels of insecticide residues (Table 3). In fact they are extremely low when compared with the value reported in USA, Canada, and U.K. (Edwards, 1976). On the other hand, water samples collected from Jaffna wells and those from tanks located in Anuradhapura (Maha Illuppallama, Tissawewa and Nuwarawewa) Polonnaruwa(Parakrama samudra and Giritale) and Minneriya did not contain any DDT, but some samples had traces of BHC, dieldrin, heptachlor and endosulfan.

Organochlorine insecticides are all of very low water solubility so that residues reported in the surveys of waters are those present in sediments (Barthel, 1966) and suspended particulate matter (Lammers, 1968). Generally the concentrations found in water are in the parts per billion (ppb) range and analysis is affected by absorption of the insecticides by materials with which the samples come in contact (e.g. glassware, plastic ware etc). Furthermore, the procedures for the analysis of soil and water have not been standardized (Frehse, 1971). Therefore, until a more systematic survey involving a large number of representative samples is carried out no firm conclusions as far as organochlorine residues in soil and water could be made.

Analysis of a few human adipose tissues taken at random from patients in Kandy gave DDT residues ranging from 10 to 102 ppm (Table 7). These are high compared to the probable world average of 5 to 10 ppm stipulated by Edwards (1976). The maximum values (indicated within brackets) reported in surveys conducted in U.K. (9.6 ppm), U.S.A. (56 1 ppm), Canada (6.8 ppm), Israel (40.6 ppm), Australia (3.5 ppm), and India (23.9ppm) are lower than the maximum obtained in Kandy. It is interesting to note that levels of 38 to 647 ppm DDT were recorded in the fat of men working between 11 and 19 years in a DDT manufacturing plant without any demonstrable ill-effects (Laws and Biros, 1967). More samples collected from different parts of the island need to be analysed to determine whether the high value recorded is normal or not. The high value obtained could be due to the fact that during the period DDT was used to control malarial mosquitoes these people were exposed to appreciable quantities of DDT.

Two possible reasons could be given for the presence of organochlorine residues in vegetables and tree crops. One, the plants could have taken up these from the soil which would have contained the residues as a result of earlier agricultural and vector control usage. Two, from the direct application of these insecticides to the crops by farmers who obtained them from the black market which existed after the ban.

The observed low levels of insecticide residues could be due to the rapid degradation of even the more persistent insecticides under tropical conditions. Although the residue levels are low, over a long period, low concentration of residues can gradually increase as they pass up a food chain as illustrated in the classical example of Clear Lake Affair in California where fish eating birds at the end of the food chain were killed (Mellanby, 1970). One consoling feature is that during the 25 years man has been exposed to these chemicals there is little evidence of resultant illness. However, according to Woodwell (1967) the spread of these persistent chemicals into all parts of the environment must be viewed with caution until we know more about their persistence, distribution and possible long-term ecological effects.

The survey in Sri Lanka seems to indicate that, unless the ban on the agricultural use of the more persistent organochlorine insecticides is lifted, there is no real danger of residue problems from this group of insecticides. A monitoring programme should however, be maintained particularly in respect of export items to maintain the residue standards set by the importing countries.

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Table 1—Samples of vegetables collected on a district basis.

District	*No. of Samples*
Anuradhapura	12
Badulla	17
Bandarawela	9
Colombo	20
Jaffna	90
Kandy	37
Kurunegala	10
Matale	17
Matara	14
Nuwara Eliya	32
Polonnaruwa	9
Trincomalee	27

Table 2—Organochlorine insecticide residues in vegetables grown in Sri Lanka (1974-1978)

Sample	*No. of Samples*	*Range of values (ppm)*					
		*DDT**	*Lindane (γ BHC)*	*Dieldrin ***	*Endrin*	*Heptachlor*	*Endosulfan****
Leafy vegetables	49	0.004–0.408 (29)	0.001–0.076 (20)	0.001–0.233 (12)	0.006–0.340 (2)	0.001–0.031 (6)	0.003–0.384 (20)
Leguminous vegetables	38	0.006–0.404 (32)	0.001–0.701 (18)	0.002–0.040 (13)	0.003–0.050 (4)	0.002–0.072 (5)	0.003–0.009 (4)
Root and tuber vegetables	60	0.003–0.609 (33)	0.001–0.029 (24)	0.001–0.122 (21)	0.005–0.027 (6)	0.001–0.009 (5)	0.001–0.188 (7)
Chillies & onions	56	0.002–1.270 (38)	0.001–0.082 (23)	0.002–0.051 (21)	0.008–0.090 (3)	0.003–0.199 (10)	0.004–0.082 (15)
Other vegetables	91	0.002–2.387 (47)	0.001–0.090 (40)	0.001–0.044 (15)	0.001–0.413 (3)	0.001–0.023 (13)	0.001–0.154 (21)

* DDT, DDD, DDE, Singly or in any combination.
** Aldrin and dieldrin, singly or in combination expressed as dieldrin.
*** Includes α and β Endosulfan.
Figure inside the brackets refer to the number of samples in which the particular insecticide residue was detected.

Table 3—Organochlorine insecticide residues in samples collected from Jaffna–1978

Sample	No. of Samples	Range of values (ppm) DDT*	Lindane (γ BHC)	Dieldrin**	Endrin	Heptachlor	Endosulfan***
Leafy vegetables	8	0.010–0.408 (5)	0.001–0.031 (7)	0.006 (1)	n.d.	0.001 (2)	0.007–0.059 (4)
Leguminous vegetables	5	0.009–0.041 (5)	0.002–0.027 (5)	0.003–0.005 (3)	n.d.	0.002–0.021 (2)	0.009 (1)
Root and tuber vegetables	12	0.003–0.072 (4)	0.001–0.011 (9)	0.001–0.021 (8)	n.d.	0.001–0.005 (4)	0.002 (1)
Chillies and Onions	15	0.004–0.206 (10)	0.003–0.039 (11)	0.001–0.038 (9)	n.d.	0.005–0.014 (5)	0.005–0.064 (4)
Other vegetables	18	0.006–0.060 (7)	0.002–0.029 (11)	0.007–0.040 (2)	n.d.	0.002–0.013 (3)	0.004–0.012 (4)
Fruits	6	0.013 (1)	0.002–0.008 (6)	0.002 (1)	n.d.	0.002 (1)	0.001-0.006 (2)
Milk	3	0.007 (2)	n.d.	n.d.	n.d.	n.d.	n.d.
Water	10	n.d.	0.0001–0.0005 (2)	n.d.	n.d.	n.d.	0.00027 (2)
Soil	8	0.012-0.203 (5)	0.002–0.003 (4)	0.004–0.032 (2)	n.d.	0.0008–0.0212 (5)	n.d.

*, **, *** See foot note of Table 2.   n.d. – not detected.

Table 4—Organocholrine insecticide residues in hulled rice grain

		ppm					
Location	*Variety*	*DDT**	*Lindane* (γ BHC)	*Dieldrin***	*Endrin*	*Heptachlor*	*Endosulfan* ***
Bombuwela ...	Bg 11–11	0.159	0.038	0.004	0.022	0.050	n.d.
	Bg 34–6	0.013	0.043	0.005	n.d.	0.030	n.d.
	Bg 34–8	n.d.	0.024	n.d.	n.d.	0.018	0.013
	H 4	n.d.	0.027	n.d.	n.d.	0.022	n.d.
	Suduru samba	n.d.	0.026	n.d.	n.d.	0.048	n.d.
Bathalagoda ...	Bg 3-5	0.031	0.016	n.d.	n.d.	n.d.	n.d.
	Bg 11–11	0.035	0.035	n.d.	0.013	0.057	n.d.
	Bg 12–1	0.012	0.011	n.d.	n.d.	n.d.	n.d.
	Bg 34–8	0.020	0.021	0.002	n.d.	n.d.	n.d
	Bg 90–2	0.009	0.019	n.d.	n.d.	n.d.	n.d.
	Bg 94–1	0.020	0.030	n.d.	n.d.	n.d.	n.d.
Maha Illuppallama ...	Bg 12–1	0.010	0.032	0.150	0.027	n.d.	n.d.
	Bg 34–8	0.007	0.031	0.161	n.d.	n.d.	n.d.
	Bg 90–2	0.006	0.024	0.106	n.d.	n.d.	n.d.
...	Bg 94–1	0.007	0.031	0.145	n.d.	n.d.	n.d.
	Bg 276–5	0.006	0.029	0.131	n.d	n.d.	n.d.

*, **, *** See foot note of Table 2. n.d.-Not detected.

Table 5—Organochlorine insecticide residues in processed products of the Marketing Department

Commodity examined	*Range of Values (ppm)*					
	*DDT**	*Lindane* (γ-BHC)	*Dieldrin***	*Endrin*	*Heptachlor*	*Endosulfan****
Canned pineapple ...	0.002–0.016	0.001–0.030	0.001	0.002	0.001–0.002	0.001–0.004
Passion Juice	o.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Beli cream	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Mango slices in syrup ...	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Grape Juice cordial ...	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Golden melon (jam) ...	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Woodapple (jam) ...	n.d.	0.001	n.d.	n.d.	n.d.	n.d.
Tomato sauce	0.002	n.d.	n.d.	n.d.	n.d.	n.d.

*, **, *** See foot note of Table 2. n.d.-not detected.

Table 6—Organochlorine insecticide residues in export products of Sri Lanka

		Range of values (ppm)					
Commodity	*No. of Samples*	*DDT**	*Lindane-* (γ-BHC)	*Dieldrin ***	*Endrin*	*Heptachlor*	*Endosulfan ****
Tobacco ...	20	0.004–0.250 (15)	0.001–0.278 (9)	0.001–0.042 (6)	0.140 (1)	0.003–0.250 (3)	0.030 (1)
Canned pineapple ...	7	0.002–0.016 (2)	0.001–0.003 (4)	0.001 (1)	0.002 (1)	0.001–0.002 (2)	0.001–0.004 (3)
Cardamom ...	3	0.060 (1)	0.001 (1)	0.015 (1)	0.010 (1)	n.d.	0.005 (1)
Cocoa bean ...	3	0.010 (1)	0.002 (1)	0.002 (1)	n.d.	n.d.	0.009 (1)
Coffee bean ...	6	0.015 (1)	0.009 (1)	0.007 (1)	n.d.	0.002 (1)	0.006 (1)
Tea **** ...	29	0.005–0.161 (23)	0.005–0.029 (20)	0.005 (3)	n.d.	0.022–0.063 (3)	0.007 (4)

*, **, *** See foot note of Table 2. n.d.-not detected

**** Analysed at the Tropical Products Institute, London

Table 7—DDT residues in human fat samples (Kandy, Sri Lanka) (ppm) *

Sample No.	Sex	4,4 DDE	O,p TDE	4,4 DDD	2,4 DDT	4,4 DDT	Total DDT
1	Female	20.35	1.88	2.44	2.94	16.75	44.36
2	Male	8.70	—	—	—	6.64	15.34
3	Male	9.40	—	—	1.24	5.62	16.26
4	Male	5.18	—	0.26	—	5.25	10.69
5	Male	68.72	—	—	—	33.30	102.02

* Analysed at the Pesticide Residue Laboratory, Bureau of Plant Industry, Manila, Philippines.

Table 8 Pesticide residue tolerences recommended by FAO/WHO * (ppm)

Crop	*DDT*	*Lindane (γ-BHC)*	*Dieldrin*	*Endrin*	*Heptachlor*	*Endosulfan.*
Root and Tuber vegetables ...	1	—	0.1	—	0.2	0.2
Other vegetables ...	7	3	0.1	—	0.05	2.0
Tropical fruits	3.5	—	—	—	—	2.0
Rice	(0.2)	0.5	0.02	0.02	0.02	0.1
Milk	1.25	0.2	0.15	0.02	0.15	0.5
Tea	(0.2)	(0.2)	—	—	—	30.0

Figures in brackets refer to tolerances to residues in Japan (Tomizawa, 1977).

* Summary List of CODEX maximum limits for pesticide residues. Joint FAO/WHO Food Standards Programme Codex Committee on pesticide residues, ninth session. The Hague, 14–21 February, 1977.