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## AN ASSESSMENT OF THE DDT RESIDUE SITUATION IN AN URBAN MILIEU

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A history of DDT application for mosquito control in and around the city of Edmonton is given. Results of analyses of soil samples collected around Edmonton in 1968, 1969, and 1970 showed that the DDT level is fairly low and the top 5 inches of soil has more DDT than the next 5 inches below it. Levels in the later years were lower. Subsequent analyses from the same samples after treatment with DDT-dehydrochlorinase showed that most gas chromatograph peaks originally thought to represent DDT were in fact caused largely by other materials probably PCB's.

Nous présentons un compte rendu sur le control des moustiques par l'application du DDT autours et dans la ville d'Edmonton. L'analyse des résultats des prélèvements de sol collectionnés autours d'Edmonton en 1968, 1969, et 1970 a démontré que le taux du DDT était passablement bas et qu'il y avait plus de DDT entre la surface et 5 pouces (12.5 cm) de profondeur qu'entre 5 pouces (12.5 cm) et 10 pouces (25 cm) de profondeur. Les taux de ces dernières années était plus bas. Après avoir soumis ces mêmes prélèvements au DDT-dehydrochlorinase d'autres analyses ont démontré que la pluspart des apogées de l'analyse chromatographique au gaz ne représentaient pas comme on l'a pensé le DDT mais plustôt d'autres composés biphenyl polychlorinés.

The mosquito problem in the city of Edmonton, Alberta has been, for a long time, the concern of the city Parks and Recreation Department, and the Provincial Departments of Health and of Agriculture, with the Department of Entomology at the University of Alberta acting in an advisory capacity. Through the cooperation of these departments a mosquito control program was started in 1953 and DDT was being used almost exclusively for this purpose until 1969.

In the last few years, investigations have centered on the possible threat to biota created by the accumulation of long-lasting chlorinated hydrocarbon residues in the soil and in food chains. This investigation was aimed at securing data on the accumulation of DDT in the soil and, if possible, on its movement through the soil.

A summary of the formulations, dosages, and amounts of DDT used and the areas treated in and around the city of Edmonton is given in Table I and the map in Fig. 1 shows the total amount of DDT (active ingredient) per acre applied to different parts of the city mosquito control area since 1953. In almost all years those areas that contained standing water as well as low-lying dry areas that could contain water were sprayed. Table 2 shows the total amount of the different DDT formulations used and it also shows that over ten tons of actual DDT have

1. Deceased.

been used since 1953.

Soil samples were taken at a number of locations within the city of Edmonton mosquito control area (Fig. 2). At each location, 3 pairs of cores were taken and the analyses were done on the top 5 inch core and the 5-10 inch depth core separately. Samples of 100 or 50 g from each core were used for extraction following Tyo's method (unpublished) which can be summarized as follows:

- Blend 100 g of soil and 175 ml 10% acetone-90% acetonitrile in a Waring Blender at high speed for 5 minutes.
- Centrifuge the mixture for 5 minutes at 1500 rpm and decant the solvent into 2000 ml separatory funnel containing 1000 ml water saturated with sodium sulfate and 100-125 ml of redistilled petroleum ether.
- To the sediment remaining after centrifugation add 100 ml acetonitrile and mix well, centrifuge and decant the solvent into the separatory funnel.
- Shake the funnel vigorously for one minute and add aqueous sodium sulfate to bring the total volume to approximately 1850 ml. Again shake the funnel vigorously for five minutes and vent as required.
- Allow the separatory funnel to stand until phase separation is complete, drain and discard the bottom layer. Rewash the top layer with 400-500 ml of aqueous sodium sulfate solution. Allow layers to separate and again discard the lower phase.
- Drip the petroleum ether from the separatory funnel through a powder funnel containing 1.5-2 inches anhydrous sodium sulfate into a 400 ml beaker. Wash separatory funnel with petroleum ether.
- Evaporate the petroleum ether extract until the volume is reduced to 40-50 ml.
- Place the extract on a chromatographic column containing activated florisil and topped with anhydrous sodium sulfate and elute with 150 ml of petroleum ether, followed by 200 ml of 6% diethyl ether (redistilled)-94% petroleum ether.
- Evaporate the final eluate from the column to near dryness, transfer with benzene to a final volume of 1 or 2 ml.

A Varian gas chromatograph 1200 with a 3 mm  $\times$  90 cm long Pyrex glass column and a 250 mc Tritium foil detector was used for the analysis. The column packing used was 6% Q.F. - 1 and 4% SE - 30 mixed silicons on 60/80 chromosorb W, and the recorder was Disc Integrator model 224-4. The temperature of the column, injector and detector were 190 C, 185 C, and 200 C respectively and the nitrogen carrier flow at approximately 40 ml/min.

To estimate the recovery of this method and that of the florisil, a series of extractions and analyses were undertaken on soil with or without known amounts of DDT. The results indicated that 91% recovery of the DDT could be attained.

The water contents of the soil samples analyzed were also determined to permit estimation of the amount of DDT on a dry weight basis.

#### **RESULTS AND DISCUSSION**

Caution must be exercised in evaluating gas chromatograms since naturally occurring compounds may give peaks with the same retention time as the insecticide sought. Although it is not conclusive evidence, the characteristics of the patterns of DDT, DDE and o, p-DDT peaks, which were observed quite frequently in the chromatograms, were assumed to represent the actual insecticides.

Table 3 summarizes the actual p, p'-DDT residue levels obtained in soil samples analyzed. The results indicated that the levels of DDT encountered are generally fairly low and in almost all samples the upper 5 inch core of soil has more DDT than the lower 5 inch core. The results

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also show a decrease in the DDT level in 1970 in almost all locations which were not sprayed in 1969. [Small amounts of material with retention times of DDE and DDD were found but these were not quantified and are not reported here.]

The DDT level in the soil around Edmonton is very low in comparison with some other places which have been using DDT for a similar time (Voerman and Besemer, 1970; Saha and Sumner, 1971; Wiersma et al., 1971). Dixon (1969) reported that the DDT residue levels in the soil around Winnipeg, which had been treated with 1 lb DDT per acre for five years prior to his analyses, ranged from 3.62 to 29.9 ppm. Brust (1971) reported similar residue levels in soil and surface vegetation in the Winnipeg area. Duffy (personal communication) found that the DDT residue level of an area at Fort Churchill, Manitoba, which had been sprayed intermittently at rates up to 0.25 lb. DDT/acre for about 12 years, ranged from 0.05 to 1.98 ppm.

For the 17 locations sampled in 1968 the correlation between the amount of DDT applied to the area and the amount detected in the soil was about 0.12. The same correlation for the 1970 data was about 0.34. This suggests either an unexpected degree of mobility of DDT through the soil, or that some of the levels found represent some other material, or both.

To test the latter possibility, extracts of the same Edmonton soil samples were treated with the enzyme DDT-dehydrochlorinase using method of Gooding et.al., (1972) prior to gas chromatography with the results shown in Table 4. It is clear from this table that a major part of most of the peaks originally interpreted as DDT, does not in fact represent this material. Attempts to specifically determine this (these) facsimile material(s) have not as yet been successful, but there is some reason to believe that they may be polychlorobiphenyls (PCB's). Further pursuit of their identity is likely to prove time consuming and expensive and is outside the scope of this study. A few further samples were taken and assayed with a view to filling in gaps in the distribution pattern and confirming the original figures. The clean-up method outlined by Reynolds (1969) was tried with a view to separating PCB's from DDT but with less success than he obtained.

This finding modifies the earlier results in the sense that true values of DDT residues in Edmonton soil are lower, in most samples substantially lower, than at first indicated. In short, there is no DDT residue problem, as at present understood, in Edmonton.

#### ACKNOWLEDGEMENT

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Table 1. History of DDT application for mosquito control in and around the city of Edmonton, Alberta.

Year	Kind of Control	Area treated in acres	Formulation	Dosage DDT	Amount of DDT used*	Area treated outside city limits
1970		NO DDT WAS	S USED			
1969	Aerial	9046	2.5% DDT in bentonite	0.05 lb/ acre	482.3 lb	3 miles north (east ½) and 2 miles east and south
	Ground	135	3% DDT in diesel oil	0.24 lb/ acre	32.4 lb	
		26	Tossits	1 tossit/ 750 sq ft	1515 tossits	
	Fogging		100% technical flakes + diesel oil		165 lb	
1968	Aerial	14225	2.5% DDT in bentonite	0.05 lb/ acre	711.3 lb	12 miles west and 2 miles east, north and south
	Ground	996	2.5% DDT in bentonite	0.05 lb/ acre	49.8 lb	
		222	3% DDT in diesel oil	0.24 lb/ acre	53.3 lb	
		30.5	Tossits	1 tossit/ 750 sq ft	1764 tossits	
1967	Aerial	9650	2.5% DDT in bentonite	0.05 lb/ acre	482.5 lb	12 miles west and 3 miles east, north and south
	Ground	450	3% DDT in diesel oil Tossits	0.24 lb/ acre 1 tossit/	108.0 lb 1710	
	Fogging		100% technical flakes	750 sq ft	tossits 50.0 lb	

Table 1. (continued). History of DDT application for mosquito control in and around the city of Edmonton, Alberta.

Year	Kind of Control	Area treated in acres	Formulation	Dosage DDT	Amount of DDT used*	Area treated outside city limits
1966	Aerial	17250	2.5% DDT in bentonite	0.05 lb/ acre	862.5 lb	3 miles north, east, and south. 3 miles north west to 12 miles south west.
	Ground	2990	3% DDT in diesel oil Tossits	0.24 lb/ acre 1 tossit/ 750 sq ft	717.0 lb 4500 tossits	
1965 <sup>.</sup>	Aerial	15000	2.5% DDT in bentonite	0.05 lb/ acre	750.0 lb	12 miles west and 2 miles east, north and south.
		6000@	2.5% DDT in bentonite	0.05 lb/ acre	300.0 lb	
	Ground	2766	3% DDT in diesel oil	0.24 lb/ acre	663.8 lb	
			Tossits	1 tossit/ 750 sq ft	10000 tossits	
1964	Aerial	21000	1.7% DDT in bentonite	0.05 lb/ acre	1050.0 lb	12 miles west and 2 miles east, north, and south.
	Ground	1250	3% DDT in diesel oil	0.24 lb/ acre	300.0 lb	,
			Tossits	1 tossit/ 750 sq ft	8000 tossits	
1963	Aerial	15000	5% DDT in bentonite	0.05 lb/ acre	750.0 lb	3 miles west, 1 mile north and east and 2 miles south
	Ground	2500	3% DDT in diesel oil	0.24 lb/ acre	600.0 lb	
			Tossits	1 tossit/ 750 sq ft	13600 tossits	
	Fogging		4% DDT in diesel oil		160.0 lb	
1961	Aerial	6000	5% DDT in bentonite	0.05 lb/ acre	300.0 lb	3 miles north and west and 2 miles south and east

Table 1. (concluded). History of DDT application for mosquito control in and around the city of Edmonton, Alberta.

Year	Kind of Control	Area treated in acres	Formulation	Dosage DDT	Amount of DDT used*	Area treated outside city limits
1961	Aerial	4000	3% DDT in diesel oil	0.24 lb/ acre	960.0 lb	
	Ground	1600	3% DDT in diesel oil	0.24 lb/ acre		
			Tossits	1 tossit/ 750 sq ft	13250 tossits	
1960 and 1959	Aerial	4000	3% DDT in diesel oil	0.24 lb/ acre	960.0 lb	3 miles north and west and 2 miles east and south
		6000	5% DDT in bentonite	0.05 lb/	300.0 lb	
	Ground	1000	3% DDT in diesel oil	0.24 lb/ acre	240.0 lb	
1957	Aerial	2000	5% DDT in bentonite	0.05 lb/ acre	100.0 lb	5 miles north and 3 miles south
		4300	3% DDT in diesel oil	0.24 lb/ acre	1032.0 lb	
	Ground	700	3% DDT in diesel oil	0.24 lb/ acre	168.0 lb	
			Tossits	1 tossit/ 750 sq ft	12000 tossits	
1956	Aerial	5500	5% DDT in bentonite and 3% DDT in diesel oil	0.05 lb and 0.24 lb/ acre	797.5 lb	3 miles north, south, east, and west
	Ground	4500	3 and 6% DDT in diesel oil	0.24 lb/ acre	1080.0 lb	
		2000	Tossits	1 tossit/ 750 sq ft	87150 tossits	
1955	Aerial	2600	6% DDT in diesel oil	0.24 lb/ acre	624.0 lb	3 miles north, south, east, and west
	Ground	34000	Tossits	1 tossit/ 750 sq ft	200000 tossits	anu wesi
	Fogging		5% DDT	700 oy 11	90.0 lb	

\* No information was available for 1962, 1958, 1954, and 1953.

@ Late season spraying.

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Table 2. Kind and Total Amount of DDT used in the Mosquito Control Program since 1953.

Kind of DDT	Amount used	Actual DDT used in lb.
3% DDT in Diesel oil	44998 gal	10799.5
6% DDT in Diesel oil	5415 gal	2170.2
2.5% DDT in bentonite	144334 lb	3608.4
5% DDT in bentonite	49255 lb	2462.8
1.7% DDT in bentonite	63000 lb	1050.0
100% DDT technical flakes	375 lb	375.0
5% DDT	225 gal	90.0
Tossits	353489 tossits	
TOTAL		22055.9

Table 3. DDT residues in soil around Edmonton, Alberta.

Location*	Year	ppb DDT/wet wt			ppb	ppb DDT/dry wt			
		minimum	maximum	average	minimum	maximum	average		
	10/0								
1-a	1968	6.5	34.0	15.7	9.5	47.0	22.0		
	1969	14.0	24.0	17.3	28.0	63.0	37.7		
	1970	26.2	73.0	44.6	36.3	87.0	61.0		
1-b	1968	5.5	7.6	7.3	6.4	10.6	9.0		
	1969	8.0	26.0	14.7	12.0	54.0	27.7		
	1970	4.7	6.5	5.5	6.5	11.2	8.2		
2-a	1968	13.6	33.1	22.3	16.7	54.8	32.0		
	1969	19.0	30.0	25.0	26.0	37.0	32.0		
	1970	28.0	96.0	65.0	31.0	133.0	81.6		
2-b	1968	4.8	41.4	17.1	5.9	67.3	29.7		
2-0	1969	3.0	9.0	5.3	4.0	11.0	6.7		
	1970	1.0	52.0	39.0	35.0	57.0	42.6		
	1770	1.0	52.0	57.0	55.0	57.0	42.0		
3-a	1968	8.4	49.7	27.0	11.3	71.1	37.5		
	1969	20.0	59.0	37.0	23.0	79.0	48.0		
	1970	11.0	57.0	33.3	14.0	73.0	45.7		
2.1	10/0	4 1	( )	5.0	5.0	75	6.5		
3-b	1968	4.1	6.0	5.2	5.2	7.5			
	1969	11.0	33.0	18.7	15.0	47.0	25.7		
	1970	9.0	17.0	13.0	15.0	20.0	11.7		
4-a	1968	2.3	4.5	3.1	2.7	5.2	3.7		
4-b	1968	1.3	2.5	1.9	1.5	3.0	2.3		

Location*	Year	ppt	DDT/wet w	ppb DDT/dry wt			
		minimum	maximum	average	minimum	maximum	average
5-a	1968	8.8	533.4	191.9	11.1	720.1	257.6
5-b	1968	5.2	8.6	6.4	6.5	10.6	7.9
6-a	1968	3.7	4.7	4.1	4.7	5.4	4.8
6-b	1968	2.5	3.3	2.8	2.9	4.0	3.3
7-a	1968	2.9	70.3	32.2	4.3	106.2	48.3
7-b	1968	20.5	69.5	38.8	30.4	101.5	56.5
8-a	1968	7.7	10.0	9.1	10.0	12.4	11.5
8-b	1968	2.9	5.8	4.2	3.7	7.0	5.2
9-a	1968	0.9	3.6	2.3	1.0	4.9	3.1
9-b	1968	0.3	2.0	1.4	0.4	2.3	1.6
10-a	1968	1.9	3.6	2.8	2.4	4.5	3.5
10-ь	1968	1.5	1.9	1.8	1.8	2.3	2.1
1 <b>1</b> -a	1968 1970	6.8 1.1	7.1 2.1	7.1 1.5	8.4 1.3	9.1 4.2	8.8 2.3
11-b	1968 1970	1.5 0.6	2.2 0.8	1.8 0.7	1.8 0.7	2.7 1.0	2.2 0.9
12-a	1968	3.1	3.5	3.2	3.5	4.0	3.7
12-b	1968	1.8	7.6	3.8	2.0	8.4	4.2
13 <b>-</b> a	1968	1.8	3.6	2.8	2.2	4.6	3.6
13-b	1968	1.5	2.7	2.2	1.8	2.9	2.6
14-a	1968 1970	5.9 2.0	40.5 3.4	26.6 2.8	9.4 2.0	53.1 3.7	32.3 3.0
14-b	1968 1970	3.3 1.6	4.7 3.3	3.8 2.3	4.1 1.7	4.4 3.4	4.8 2.4

Table 3. (continued). DDT residues in soil around Edmonton, Alberta.

Location* Year		ppb DDT/wet wt			ppb DDT/dry wt		
		minimum	maximum	average	minimum	maximum	average
15-a	1968	1.2	29.3	11.3	1.7	45.1	17.4
15-b	1968	2.0	11.1	5.4	2.8	15.9	7.9
16-a	1968	2.6	23.6	12.2	3.3	30.3	15.9
	1969	9.0	21.0	14.0	12.0	30.0	19.3
	1970	2.7	4.9	4.0	4.4	7.3	6.1
16-b	1968	1.9	2.6	2.3	2.3	3.3	2.8
	1969	7.0	22.0	12.7	8.0	33.0	17.7
	1970	0.7	3.6	1.9	1.0	5.3	2.7
17-a	1968 1969	1.5	23.5	14.1	1.9	29.1	17.3
	1970	6.2	12.6	6.3	8.3	21.0	9.8
17-b	1968 1969	1.1	20.0	10.7	1.4	24.0	13.1
	1970	5.7	17.0	10.1	7.5	23.6	13.9

Table 3. (concluded). DDT residues in soil around Edmonton, Alberta.

\* a: top 5 inch core

b: 5-10 inch depth core.

Table 4. Comparison of Estimates of DDT Residues in Soil before and after Treatment withDDT Dehydrochlorinase. The numbers in the body of the table are picomoles ofDDT.

Sample No.	Initial	Analysis	After adding 0.2 µg DDT to Metabolized Samples (2)		
	Before DDT-ase	After DDT-ase	Before DDT-ase	After DDT-ase	
	treatment	treatment*	treatment	treatment <sup>**</sup>	
	(1)	(2)	(3)	(4)	
28/1	180.4	152.7	784.1	196.4	
48/2	1340.6	486.0	877.2	339.2	
21/2	985.6	680.2	1082.0	339.2	

 \* 100% conversion of DDT→DDE was achieved in control run of first DDT-ase incubation (1 μg DDT or 28,200 picomoles incubated).

\*\* 97% conversion of DDT→DDE achieved in 2nd DDT-ase incubation using 564 picomoles DDT.

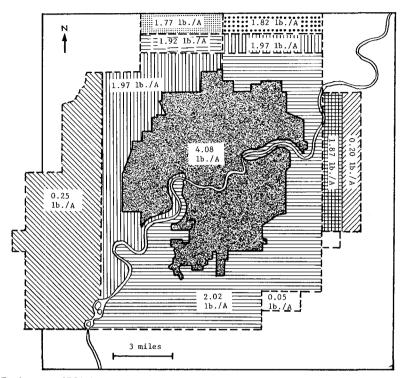


Fig. 1. Total amount of DDT (actual ingredient) per acre sprayed at different locations within the city of Edmonton mosquito control area since 1953.

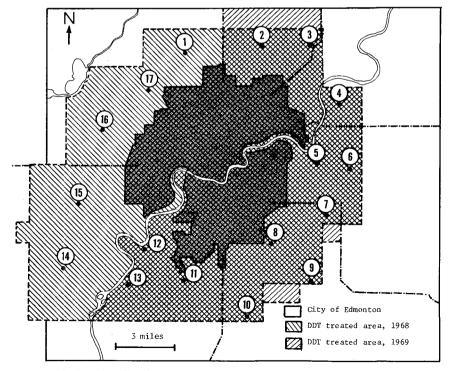


Fig. 2. Location of soil samples collected.