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BIOLOGICAL TRANSFER AND LOSS OF ³⁶Cl-LABELED DDT IN AN
OLD-FIELD ECOSYSTEM

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PART I

BIOLOGICAL TRANSFER AND LOSS OF ³⁶Cl-LABELED DDT IN AN OLD-FIELD ECOSYSTEM

Douglas J. Forsyth*

INTRODUCTION

The enclosed 10-acre old-field plot treated in June 1969, with chlorine-36 labeled DDT was sampled each year from 1969 through 1974 to monitor the fate of the insecticide in the soil and biota. In order to provide data on compartmentalization of DDT in the vegetation, invertebrates and vertebrates inhabiting the plot, sampling was carried out to estimate both body burdens of DDT and biomass of populations. Another aspect of this study, the determination of rates of accumulation of residues by invertebrates and vertebrates, has been reported previously (Forsyth and Peterle 1973; Forsyth *et al.* 1975; Peterle 1975). This report describes (a) temporal patterns of DDT residues in soil and biota from 1969 through 1974 and (b) quantities of DDT held in the soil and biotic compartments of the ecosystem.

METHODS AND MATERIALS

Collection and Analysis of Soil

In view of the uneven distribution of DDT-impregnated clay granules following application in 1969 (Bandy and Peterle 1972), the sampling program for soil did not attempt to characterize the

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residue content of the soil in the entire plot by selecting random sites for collection. Instead, a series of fixed sites was sampled annually. Soil cores collected for analysis of mite populations and soil residues by a co-operating scientist at Syracuse University were rendered useless for residue analysis by the procedure used to extract invertebrates. A small-scale reapplication of labeled DDT-treated granules was carried out in 1976 to supplement the data lost for 1969 and 1970. The movement of DDT in soil, air and water following the reapplication is described in the attached report by B. Grau.

During October of 1971 through 1974, soil cores were collected from five sites that ranged from 2309 to 8617 disintegrations per minute (dpm) in chlorine-36 activity of treated granules sampled during the application by helicopter (Bandy and Peterle 1972:219). An additional five sites were sampled in 1971 and 1974 to provide soil from 10 sites for comparison of residues between those two years. Another 20 sites in the treated plot sampled in 1971 have not been analyzed as yet, nor were these sites sampled in subsequent years due to the amount of labor involved in analysis.

Two soil cores, 5.08 cm in diameter and 6 cm deep were collected at each site, 1-2 m apart each year, except 1974 when four cores per site were collected. Samples were divided into two layers, the top 3 cm and the second 3 cm, and held frozen (-20°C) until the time of analysis. The method of analysis, consisting of triple extraction in hexane followed by determination of Cl-36 activity

in the extract by liquid scintillation spectrometry (LSS), has been described previously (Peterle 1975).

Air Sampling

A portable air sampling device was utilized to trap airborne DDT in ethylene glycol. The device was described in Bandy and Peterle (1972). In 1970 and 1971, the air sampler was housed in a plastic canopy in an effort to minimize loss of DDT vapor through air currents. Analysis of the resulting samples showed no detectable residues, however, and a new approach was taken. Plastic tubing 1.5 cm in diameter and 2 m long was attached to each of the air intakes on the air sampler and laid along the ground next to the detritus layer without disturbing the vegetation. The 1.5 m sections of the tubing that were in contact with the detritus were perforated with about four 1 mm holes per cm^2 , and the ends were plugged to prevent arthropods from entering. Air was pumped through the ethylene glycol of the sampler at the rate of 1 cubic foot per minute (0.028 cubic meters per minute) for several hours during the afternoons of 4-5 days for a total of 24 hours per sample. This procedure was adopted to obtain air samples while the ambient temperature was relatively high and conditions for volatilization of DDT would have been optimal. Two sites were sampled in 1971, and the second site was sampled again in 1972.

The ethylene glycol resulting from air sampling was extracted for DDT content by the following method. Samples were first filtered

to remove particulate matter, then washed three times with 200 ml aliquots of hexane in a separatory funnel. The pooled hexane extract (600 ml) was poured through a glass column containing 20 cm of sodium sulphate granules to remove water. Two rinses of 50 ml hexane were poured through the granules to bring out the remaining hexane extract. The extracts were flash-evaporated (45°C; 200 rpm) to several millileters and transferred, rinsing three times with hexane, into scintillation vials in which they were evaporated to 3 ml under an air-stream. Scintillation cocktail (15 ml) and Triton X-100 emulsifier (2 ml) were then added and C1-36 activity was determined by LSS.

Collection and Analysis of Vegetation

Plants were collected monthly, June through October in 1970 and 1971, and in August 1972, November 1973, and October 1974. Collections were made each time at the five sites designated for soil sampling. Six species were taken, representing the major vegetative cover of the study area: bluegrass (Poa spp.), couch grass (Agropyron repens), orchard grass (Dactylis glomerata), wild parsnip (Pastinaca sativa), wild carrot (Daucus carota), and yarrow (Achillea millefolium). Specimens were kept frozen (-20°C) until they were prepared for LSS analysis by the procedures described in Bandy and Peterle (1972).

Collection and Analysis of Invertebrates

Pitfall traps made by sinking quart-sized oil cans into the ground were placed at each of the 100 wooden stakes marking the 60-ft (18.3 m) grid system used in the small mammal live-trapping program. These traps were baited with a mixture of sugar and baker's yeast in water for 7-10 days each sampling period; when not in use they were turned upside down. The contents of all the traps were pooled into one container for a given collection period and frozen for residue analysis. These traps were used for collecting crickets, beetles, firefly larvae, millipedes and spiders.

Earthworms were collected at the five sites used for soil and plants; they were not included in sampling of biota in 1969. Approximately 20 worms per site were taken by clipping the vegetation on 0.12-0.25 m² of habitat containing earthworm castings and soaking the soil with diluted formalin (Raw 1959). The worms were frozen in water to prevent dessication until they could be prepared for LSS analysis.

Isopods and slugs were collected under logs and discarded wooden rabbit traps at 23 sites scattered around the study area. Included among these sites were the five localities selected for sampling of soil, plants and earthworms. In the case of isopods, no more than six individuals were taken per site for the pooled sample representing the whole study area. Slugs and isopods were frozen in water to prevent dessication.

Standard sample weights were 0.20 gm for earthworms and slugs, and 0.25 gm for isopods and insects. Earthworms were large

enough to require only one individual per sample; small specimens were prepared as 0.10 gm samples. Isopods and small insects were pooled, usually requiring 3-8 individuals. Specimens were chopped finely with scissors, weighed into scintillation vials, solubilized with NCS at 45°C for 18 hr, and bleached with 30% H₂O₂ (Bandy and Peterle 1972).

Collection and sample preparation of small mammals was described in Forsyth and Peterle (1973). In order to derive predictive curves for estimating total body burdens of DDT in short-tailed shrews (Blarina brevicauda) and meadow voles (Microtus pennsylvanicus), 14 voles and 15 shrews were first dissected to obtain samples of fat, brain, liver and muscle, then chopped finely with scissors. The resulting homogenates were subsampled for analysis by LSS and residues in the selected tissues from each individual were compared to whole body levels. These comparisons were expressed as regression equations that were used to predict whole body residues from fat in Blarina and from muscle in Sorex and Microtus. The regression equations developed were as follows:

(a) $Y = 1.153X - 0.03$ for Microtus muscle

(b) $Y = 1.181X + 0.37$ for Sorex muscle

(c) $Y = 0.094X + 0.902$ for Blarina fat

(d) $Y = 1.815X + 0.80$ for Blarina liver

By substituting ppm DDT in the muscle, fat or liver into the above equations, ppm in the whole carcass was obtained. Carcass

levels in Blarina were predicted from fat for all years except 1969 when liver residues were used in equation (d).

Garter snakes (Thamnophis sirtalis) were collected by hand wherever and whenever they were encountered in the study area. They were sampled for fat, liver and muscle, and notes were made on prey items found in the stomachs.

All organisms collected in the study area were duplicated with control specimens taken during the same sampling period from similar habitat at least 90-100 m outside the enclosure.

Gas Chromatography

In order to verify residues of DDT predicted by LSS analysis, a series of samples was extracted with hexane and the extracts were analyzed by both LSS and gas chromatography. The series consisted of four Blarina and four Microtus whole body homogenates, four hexane extracts of Cl-36 DDT-impregnated clay granules prepared in 1969, one earthworm and one sample of snake fat. In preparation for gas chromatography, weighed portions of tissues were ground in sodium sulphate using a mortar and pestle, then extracted in a glass chromatography column with 300 ml of hexane added to the column in three aliquots of 10 ml, followed by a fourth aliquot of 270 ml. Extracts were flash-evaporated to 2 ml, transferred to centrifuge tubes, and cleaned in a florisil column. Florisil cleanup was accomplished by adding the hexane extract to a 15 cm column of

florisil (1.2% deactivated) that had been wetted with an initial 60 ml aliquot of hexane. The extract was followed by four 10 ml and one 100 ml rinses of hexane and finally a 150 ml rinse of 15% methylene chloride in hexane. The cleaned extract was flash-evaporated to 2 ml, transferred to a centrifuge tube, evaporated to dryness (1 drop of mineral oil added to prevent loss of DDT residues) to eliminate methylene chloride, then made up to 1 or 10 ml with hexane for injection. Injections were made into a Hewlett-Packard 5730A, using a mixed phase column packing of 1% OV-210 at an oven temperature of 180°C.

When results indicated that hexane extracts contained less DDT residues detectable by gas chromatography and LSS than were detected in solubilized tissue analyzed by LSS, extracts were made with a 2:1 solution of chloroform and methanol (C/M). The C/M extracts were made using the same volumes as stated for hexane extraction. The extracts were evaporated to dryness in scintillation vials, the remaining lipid residue was solubilized in 0.5 ml of NCS, scintillation cocktail and Triton X-100 were added, and C1-36 activity was determined by LSS.

Biomass Determinations

The vegetation of the treated enclosure was sampled monthly from June through November of 1970 at 15 sites chosen each month from random number tables. Samples were made by clipping all vegetation from 0.25 m² quadrats and transporting the clipped material in plastic

bags to the laboratory where green vegetation was separated from dead material and sorted to species. Following oven-drying for 24 hr at 100°C, weights of species were recorded and the average monthly biomass of the combined species was calculated. An average water content of 80% was determined experimentally for use in converting dry weight per m² to standing crop of living vegetation. Roots were sampled by taking 15 cm cubes of soil from the centers of clipped quadrats. Soil was washed from the roots which were then oven-dried and weighed.

Populations of insects were estimated monthly from May through October 1971, by placing an open-ended plastic cylinder at random sites and removing the contained insects by means of an industrial vacuum cleaner operated by a portable generator (Barrett 1968). The area of the cylinder, with a diameter of 56.4 cm, was 0.25 m². Arthropods were transferred with plant debris from the vacuum cleaner into a large plastic bag and then into a killing bottle to which ethyl acetate was added. They were subsequently separated from the debris and stored by freezing.

Earthworms were collected by the formalin method (Raw 1959) from 10 randomly selected 0.25 m² sites each month from May through November 1971; in March, April, May, July and September 1972; and in June, September, October and November 1973. They were stored in 10% formalin.

Isopods, slugs and firefly larvae were censused by a technique described by South (1964). Each month from July through

November 1973, 10 samples of turf measuring 12 in x 12 in x 4 in (area = 930.25 cm^2) from random sites were collected, placed immediately in 20 gal (76 l) plastic garbage cans and transported to the laboratory. Turfs were placed edgewise in the garbage cans and water was added to a depth of 3 cm, followed by an increase 15-20 hr later to 15 cm. Two more additions of water were made on the third day to bring the level up to 2 cm below the upper edge of the turf. All invertebrates except earthworms found at each stage of flooding were removed and stored in 10% formalin.

Dry weights per m^2 were calculated by oven-drying at 100°C for 24 hr. Dry weight was converted to live weight by mean ($\pm 1\text{SE}$) percent moisture values of 76.3 ± 0.6 ($N=19$) for crickets, 76.7 ± 0.7 ($N=60$) for isopods, 82.3 ± 0.6 ($N=40$) for earthworms, and 86.3 ± 0.8 ($N=67$) for slugs. The percent moisture in crickets was assumed to apply to firefly larvae.

Biomass of small mammals was estimated by live-trapping from 3 July through 30 November 1970, from 4 April through 13 October 1971, and from 21 July through 23 November 1973, using standard Sherman traps. Two traps per station were set in a grid system at 60 ft (18.3 m) intervals. In 1970, traps were set in the evening and checked soon after dawn on alternate days; that is, 10-15 days per month. In 1971, the evening/dawn schedule was maintained, but trapping was reduced to periods during the first, second and fourth week each month until September when traps were set daily during

10-16 September and 7-13 October. In 1973, a schedule of trapping for six days towards the end of each month was adopted and traps were prebaited in the afternoon (left open to permit free access to bait), set at dusk, and checked between 2200 hr and 0500 hr. Mortality of animals in the traps was almost eliminated by this reduction of time in captivity. Captured animals were transported to the field laboratory adjacent to the study area (Bandy and Peterle 1972) where they were toe-clipped, weighed and examined for reproductive condition. Populations of Blarina and Microtus were calculated by the calendar of catches method (Petrusewicz and Andrzejewski 1962) which simply sums the total number of individuals known to be present throughout a period of trapping. These values were increased by 15% to account for animals avoiding the traps (Hilborn et al. 1976). Peak populations of Blarina were estimated from the data of the first month's trapping in 1970 and 1973 to avoid the influence of mortality due to live-trapping. Most of the Sorex taken in live-traps in 1970 and 1971 did not survive; hence, their numbers were estimated by the method developed by Zippin (1956) for two intervals of removal trapping.

RESULTS

Gas Chromatography

Duplicate analyses of DDT-impregnated clay granules and various tissues by LSS and GC provided an estimate of μgm DDT per

Table 1. Comparison of Cl-36 activity in NCS-dissolved tissues and hexane extracts with total μgm DDT (including metabolites) in hexane extracts. Minced carcasses of Blarina and Microtus were subsampled for the analyses.

Sample	dpm Cl-36		GC Analysis μgm DDT	$\mu\text{gm/dpm}$ in Hexane Extracts
	NCS-dissolved	Hexane Extract		
earthworm	13.3	0.0	0.10	--
snake fat	22.0	15.9	2.43	0.152
Blarina A-1	14.5	4.0	0.77	0.192
Blarina A-2	14.5	4.1	0.87	0.212
Blarina B-1	23.6	12.2	2.22	0.182
Blarina B-2	23.6	10.3	1.70	0.165
Blarina C-1	37.5	24.0	3.46	0.144
Blarina C-2	37.5	18.5	3.63	0.196
Blarina D	23.7	8.8	1.51	0.171
clay granule A	--	85.1	15.30	0.180
clay granule B	--	37.1	8.14	0.219
clay granule C	--	87.2	13.22	0.151
clay granule D	--	151.2	25.91	0.174
Microtus A	7.6	0.0	0.26	--
Microtus B	8.2	0.0	0.36	--

dpm of Cl-36 activity and demonstrated the presence in some tissues of a hexane-insoluble metabolite. The median value for ratios of μgm DDT residues per dpm Cl-36 activity for 12 samples was 0.177 (Table 1). A linear regression of μgm DDT as a function of dpm Cl-36 produced the equation $Y = .177X - 0.038$, which was used for converting dpm to μgm . When subsamples of Microtus carcass A and B (Table 1) were extracted with chloroform-methanol (C/M), the Cl-36 activities in the extracts were 7.3 and 2.8 dpm, respectively, demonstrating that residues detected as Cl-36 in NCS-dissolved samples were soluble in C/M but not in hexane. Hexane and C/M extracts of two additional earthworm samples gave similar results.

Residues in Soil and Air

When four soil samples from the study area were analyzed by hexane extraction and by duplicate C/M extraction, the Cl-36 activity in the C/M extracts ranged from 1.9 to 3.3 times the activity in the hexane extracts (mean $\pm 1\text{SE} = 2.5 \pm 0.3$). As all soil residue data were derived by triple hexane extraction, they have been corrected by the factor 2.5.

The top 6 cm of soil from 10 sites in the study area (Table 2) contained DDT residues averaging 1.71 ± 0.44 ppm (N=18) in 1971, compared to 1.17 ± 0.25 ppm (N=19) in 1974. The difference between years is not statistically significant ($t = 1.072$). The decline in soil residues between 1969 and 1974 was 47.3%.

Table 3. Residues of DDT in soil cores from five sites in the study area. Cores were 6 cm deep, including detritus. Data are given for 2-4 samples per site. Values for 1979 were obtained by assuming all DDT in the clay granules collected in petri dishes during application was incorporated in a soil core of 80 g dry weight.

Sampling Site*	DDT residues, ppm dry weight				
	1969	1971	1972	1973	1974
205	1.33	0.35 0.40	0.80 1.10	0.13 2.50 0.23 0.50	0.38 1.25
250	2.20	0.05 0.20	0.20 0.43	0.55 1.30 4.43 1.23	1.33 1.15
386	2.70	3.53 0.38	1.20 0.40	0.60 0.73 1.38 3.98	0.48 0.83
279	3.75	6.78 2.05	N.S.C. N.S.C.	5.30 4.50	4.05 2.44
352	4.96	4.98 3.28	7.13 7.05	1.58 2.98 1.90 4.40	2.90 1.93
Mean ppm**	2.80	1.65	2.29	1.78	1.28
SE	0.77	0.69	1.05	0.37	0.29

* Locations of petri dish samplers (Bandy and Peterle, 1972:219).

** Site 279 omitted from mean because samples were not collected (N.S.C.) in 1972.

Table 2. Comparison of DDT residues in the top 6 cm of soil from ten sites in the study area collected in 1971 and 1974. The 1969 residues represent deposition of granules on the surface area of a soil core.

Year	DDT residues, ppm dry weight			
	Mean	N	Range	SE
1969	2.22	10	0.04-4.96	0.53
1971	1.71	18	0.05-6.78	0.44
1974	1.17	19	0.01-4.05	0.25

Table 3 shows the high level of variability in soil residues within single site collections. For example, in 1973 residues at site no. 250 ranged from 0.55 to 4.43 ppm in four samples collected within 2 m of one another. Also evident in the Table is the lack of any trend toward declining soil residues from year to year. Average levels in 1972 and 1973 were both higher than those of 1971.

In 1975, an experiment was conducted to determine how deep into the soil profile DDT residues had penetrated. Results indicated (Table 4) that residues found in the second 3 cm were 0-22% of the

Table 4. Residues of DDT at four depths in soil cores from four sites in the study area.

Sampling Site	DDT residues, ppm dry weight				6-9 cm as % 0-6 cm
	0-3 cm	3-6 cm	6-9 cm	9-12 cm	
87	0.53	0.00	0.01	0.00	2.4
250	1.52	0.09	0.01	0.00	0.6
279	3.24	0.07	0.01	0.00	3.5
352	4.18	0.92	0.18	0.01	0.3

levels present in the top 3 cm. Very little penetration occurred beyond 9 cm; residues in the third 3 cm were 0.3-3.5% of those in the first 6 cm. Therefore, analysis of the top 6 cm of soil accounted for an average of 98.3% of the DDT present in soil.

Air samples collected at the surface of the detritus layer indicated (Table 5) that the amount of volatilization of DDT residue was proportional to the amount present in the soil.

Table 5. Concentrations of DDT detected in ethylene glycol through which air was bubbled from the surface of the detritus layer of two sites in the study area in 1971 and 1972. Soil levels of DDT were estimated from granule deposition. Temperatures next to the detritus layer at the beginning and end of sampling intervals were averaged.

Sampling Site	Year	Mean Temp., °C	ppm DDT in Soil	µgm DDT in eth. glycol
A	1971	15.6	5.7	1.0
B	1972	16.7	23.4	4.9
B	1972	26.1	23.4	10.6

Granule deposition indicated a soil level of 23.4 ppm in the top 3 cm at site B, which was four times the amount estimated for the soil of site A. The DDT detected at 16.7°C at site B was 4.9 times the amount detected at 15.6°C at site A. There was 2.2 times as much DDT detected at 26.1°C compared to 16.7°C at site B.

Residues in Vegetation

A trend of increasing concentrations of DDT in plants from the five sites sampled was evident from 1969 through 1974 (Fig. 1).

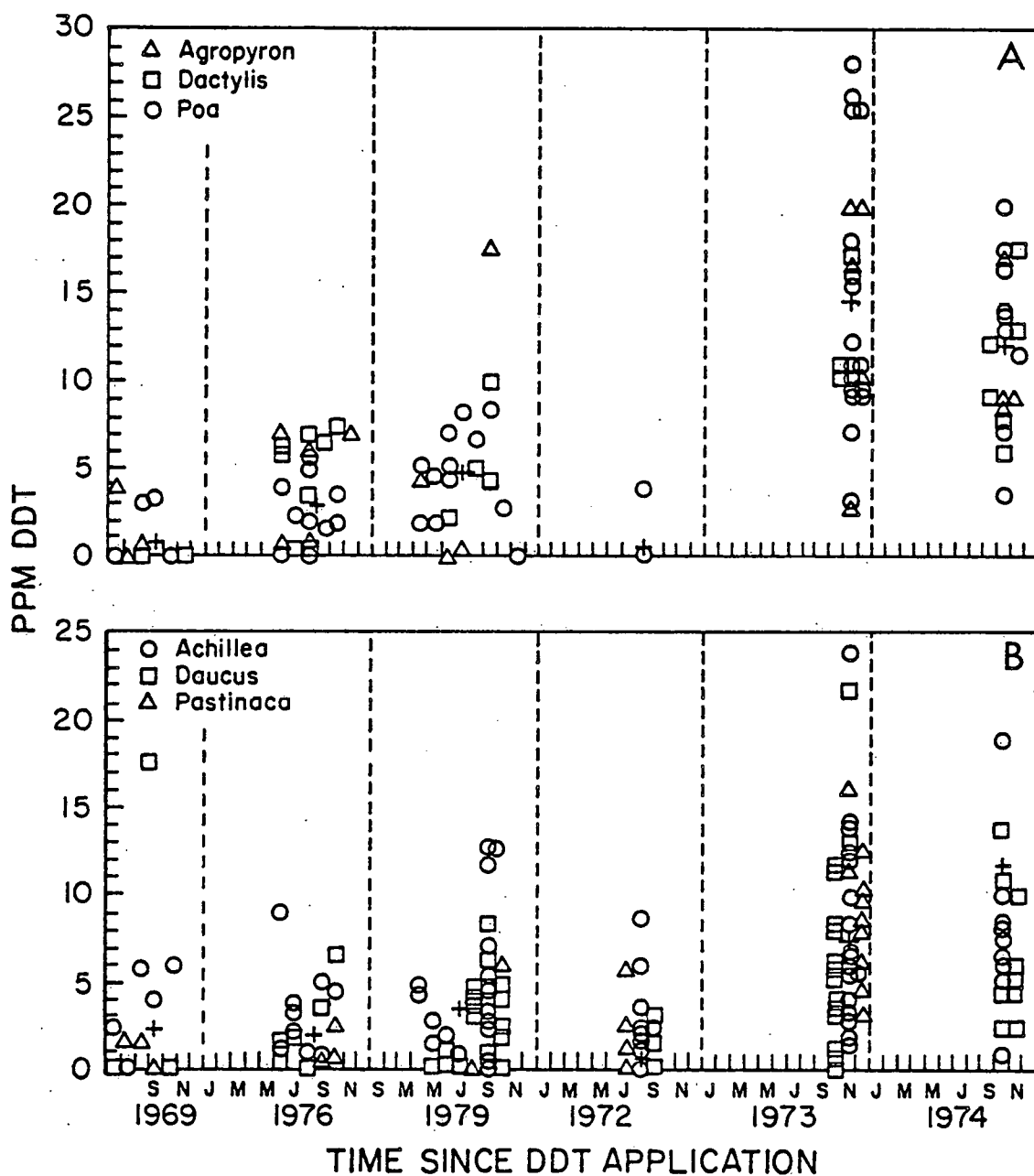


Figure 1. Temporal trends in the DDT residues of the leafy portions of three grasses (A) and three herbs (B). Circles, squares and triangles represent data for individual samples; plus (+) signs are the means for all samples each year.

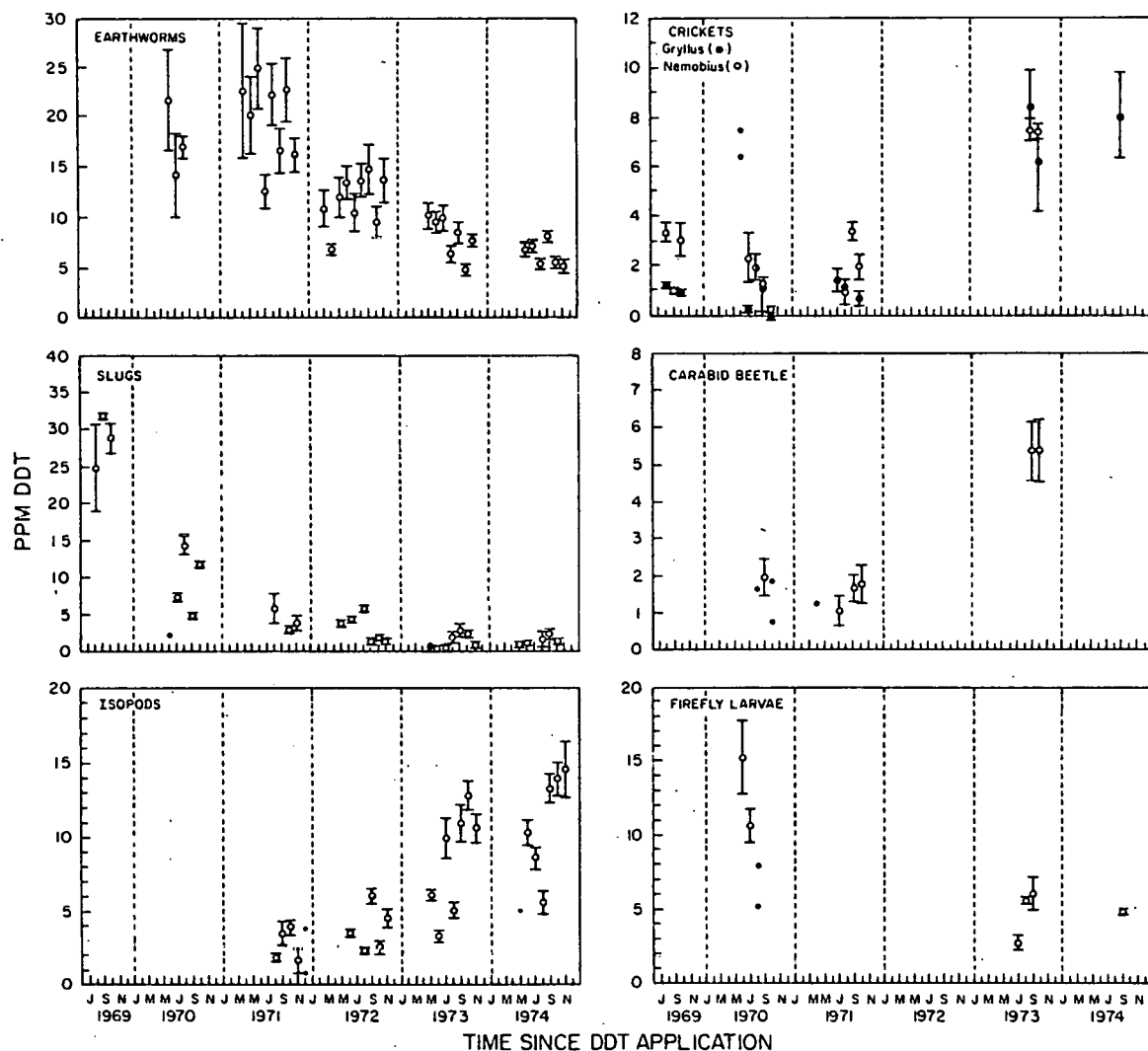


Figure 2. Temporal trends in the DDT residues of invertebrates inhabiting the soil and detritus layer. Circles and vertical bars represent means \pm 1 SE.

This trend was interrupted temporarily in 1972 when an abrupt decline in concentrations occurred. Of the six species monitored, there were no marked inter-species differences (Table 6). The herbs (Daucus carota, Achillea millefolium and Pastinaca sativa) ranged from 0.26 to 3.03 ppm in 1970 and had increased to 6.56-8.03 ppm in 1973. Grasses (Poa spp., Dactylis glomerata and Agropyron repens) accumulated slightly heavier residue burdens than herbs, increasing from 2.10 to 3.67 ppm in 1970 to 12.75 - 15.20 in 1973.

Residues in Invertebrates

The species of invertebrates that could be collected in sufficient numbers to permit residue analysis are represented in Fig. 2. Residues in all species shown, with the exception of earthworms, are considered to be representative of the entire study area, as samples were collected at sites scattered throughout the enclosure. Earthworms were collected at the five sites sampled for soil and plants.

Residues in earthworms (Lumbricus terrestris) declined from a high of 25 ppm in April 1971 to a low of 5 ppm in November 1974. There were no consistent seasonal patterns of DDT accumulation. Slugs (Deroceros larvae) attained a peak of 32 ppm in 1969, during the third month post-application. Residues declined to 5-15 ppm in 1970 and appeared to have reached a plateau in decline by 1973, as levels in that year and the next were about 1.5 ppm. Isopods

Table 6. Residues of DDT in vegetation from five sites in the study area during 1970-1974. Data given as means \pm 1 SE; sample sizes in parentheses.

Species	Year				
	1970	1971	1972	1973	1974
Herbs					
<u>Daucus carota</u>	3.03 \pm 1.38 (4)	2.85 \pm 0.57 (18)	0.27 \pm 0.20 (18)	6.56 \pm 1.12 (20)	7.05 \pm 1.3 (10)
<u>Achillea millefolium</u>	2.56 \pm 0.65 (7)	4.75 \pm 0.84 (20)	1.50 \pm 0.51 (19)	8.03 \pm 1.19 (20)	8.35 \pm 1.5 ^a (10)
<u>Pastinaca sativa</u>	0.26 \pm 0.16 (4)	1.55 \pm 1.55 (4)	0.79 \pm 0.31 (20)	7.75 \pm 0.70 (20)	D.N.A.*
Grasses					
<u>Poa spp.</u>	2.10 \pm 0.64 (8)	4.74 \pm 0.86 (11)	0.96 \pm 0.96 (4)	15.20 \pm 2.00 (20)	13.06 \pm 1.75 (9)
<u>Dactylis glomerata</u>	3.52 \pm 1.42 (5)	5.29 \pm 1.27 (5)	N.S.C.	12.75 \pm 2.30 (3)	11.02 \pm 0.4 ₁ (6)
<u>Agropyron repens</u>	3.67 \pm 1.68 (4)	5.63 \pm 4.12 (4)	0.00 (2)	14.10 \pm 3.35 (5)	11.05 \pm 1.9 (4)
Combined Species	2.50 \pm 0.42 (32)	4.09 \pm 0.46 (62)	0.86 \pm 0.21 (63)	9.76 \pm 0.73 (88)	11.99 \pm 1.0 ⁻ (39)

* D.N.A. data not available; N.S.C. no samples collected.

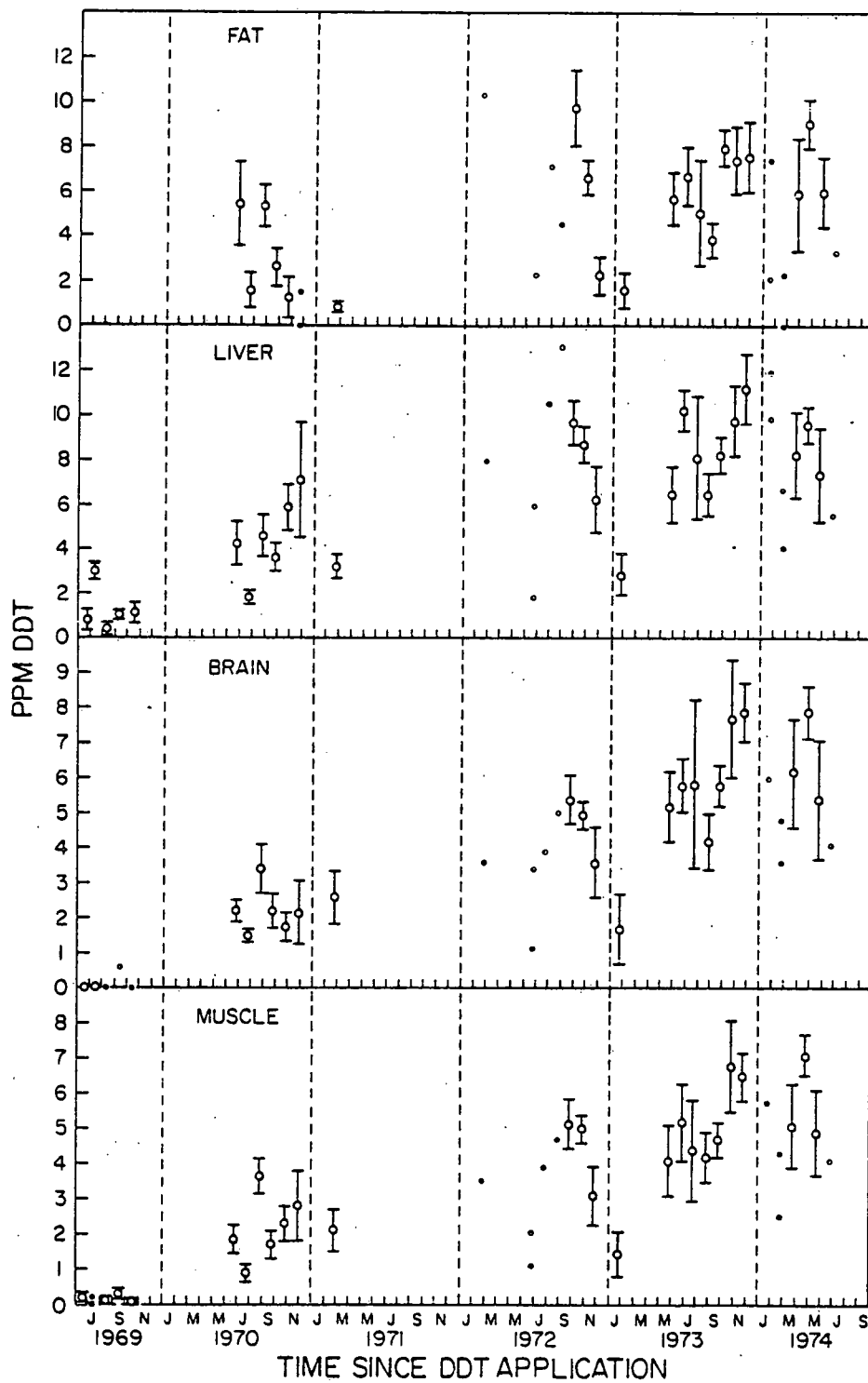


Figure 3. Temporal trends in DDT residues of four tissues of the meadow vole, *Microtus pennsylvanicus*. Open circles and vertical bars are means ± 1 SE; closed circles are individual samples.

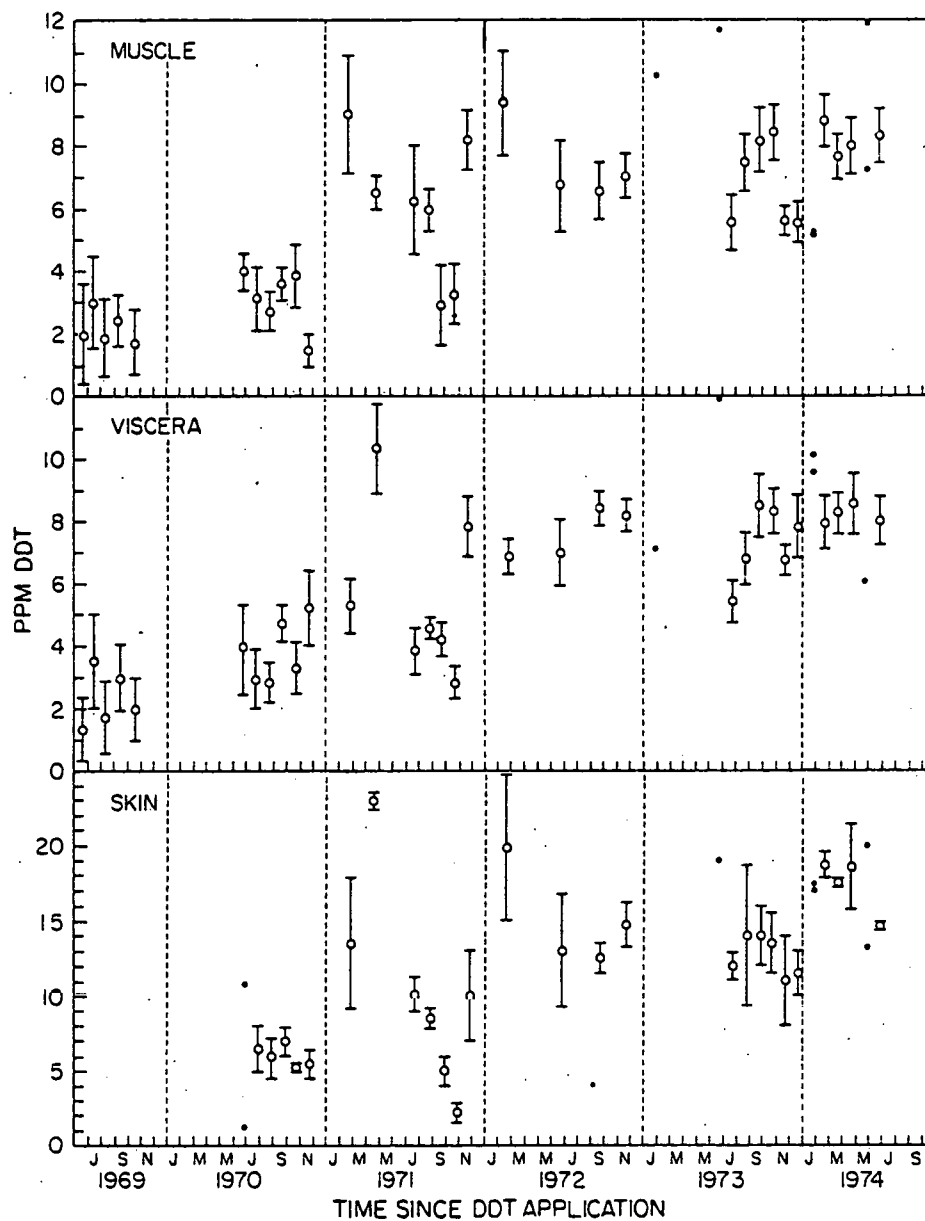


Figure 4. Temporal trends in DDT residues of three tissues in the masked shrew, *Sorex cinereus*. Open circles and vertical bars represent means ± 1 SE; closed circles are individual samples.

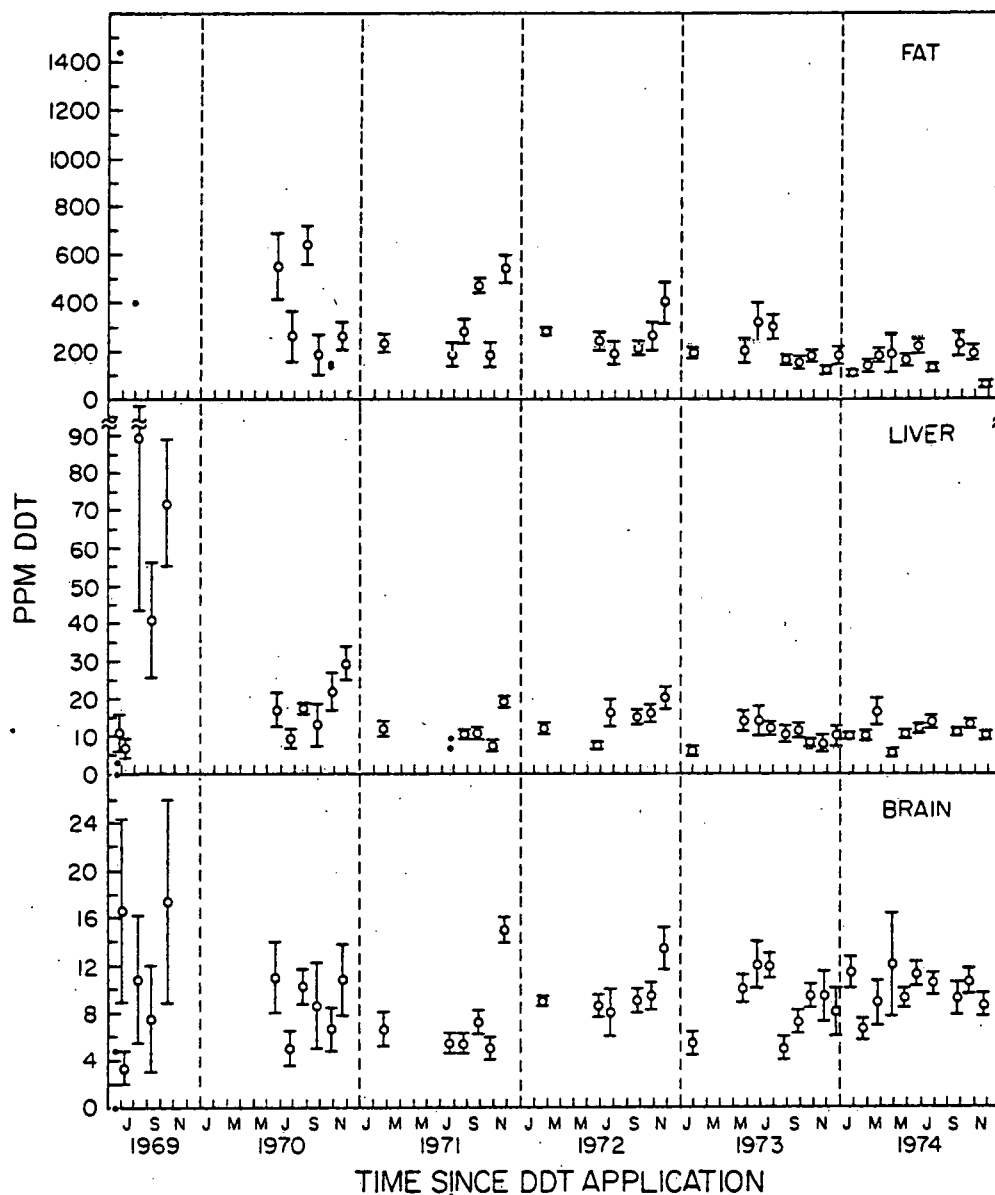


Figure 5. Temporal trends in DDT residues of three tissues in the short-tailed shrew, *Blarina brevicauda*. Open circles and vertical bars represent means ± 1 SE; closed circles are individual samples.

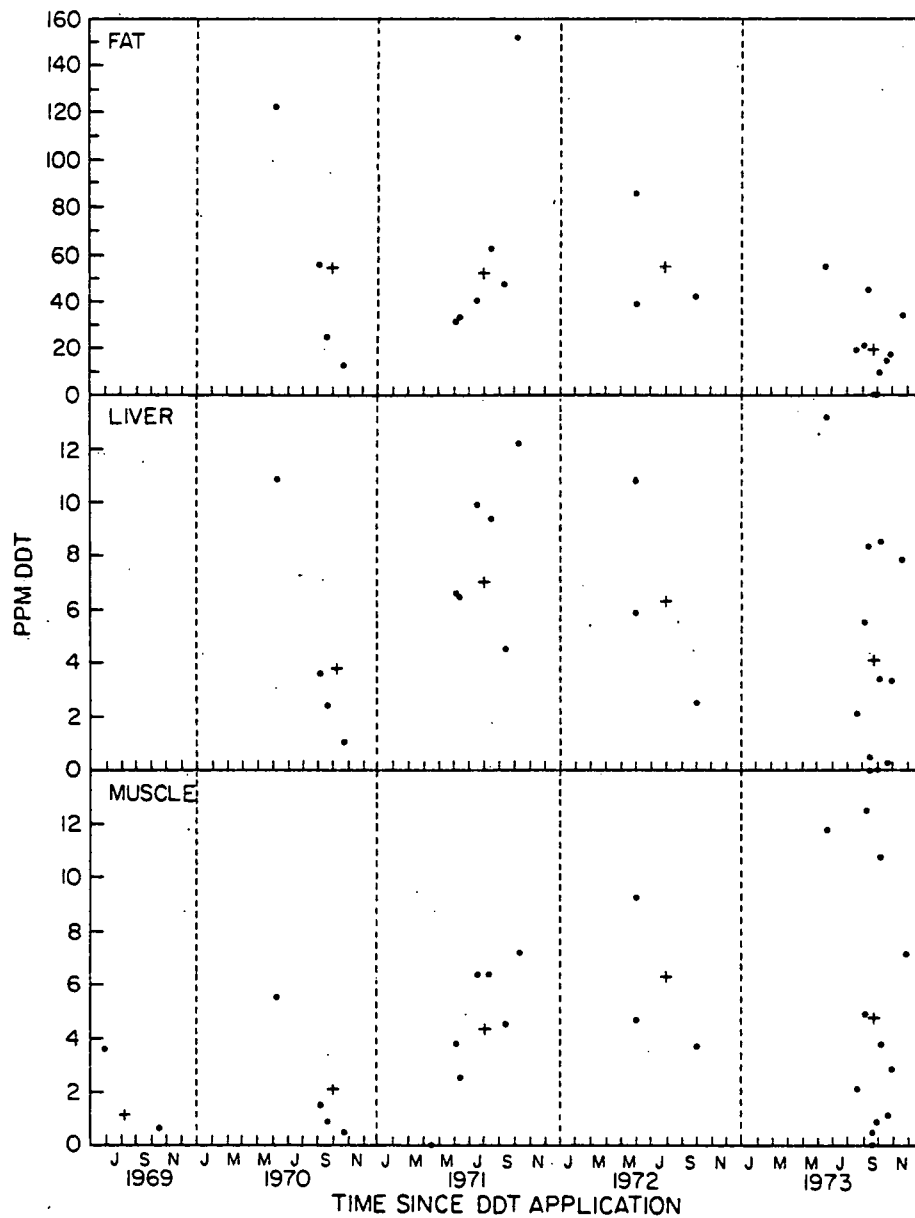


Figure 6. Temporal trends in the DDT residues of three tissues in the garter snake, Thamnophis sirtalis. Each point represents one sample. Plus (+) signs are the means for all samples each year.

(Tracheoniscus rathkei) were almost completely extirpated by the DDT application. When they became abundant enough in 1971 to permit collection, their residues averaged about 2.5 ppm. They increased to 4 ppm in 1972 and were 5.5 to 14.5 ppm in 1974. Concentrations in isopods tended to be greater in the autumn than in spring and summer.

The two species of crickets, Gryllus pennsylvanicus and Nemobius allardi, differed very little in residue content. Concentrations increased from about 1.5 ppm during 1969-1971 to about 7 ppm in 1973 and 1974. Similarly, the carabid beetle, Abacetus permundus, exhibited a trend of increasing DDT residues that averaged 1.5 ppm in 1970 and 5.4 ppm in 1973. Residues in larvae of the firefly (Photinus pyralis) declined from a high of 15.3 ppm in June 1970 to 4.5 ppm in 1973 and 4.8 ppm in 1974.

Residues in Vertebrates

Tissues of the vole, Microtus pennsylvanicus, displayed a pattern of increasing residue concentrations that reached a maximum in 1973 and remained at similar levels in 1974 (Fig. 3). Liver contained higher levels than fat, brain or muscle, attaining a peak mean level of 11.3 ppm in November 1973. Residues in fat and brain were about 7.5 ppm in November 1973. Muscle contained slightly less at 6.5 ppm.

The smaller of the two shrews, Sorex cinereus, also underwent an increase in DDT that appeared to plateau in 1972 (Fig. 4). Muscle

and viscera reached similar plateau levels of 7-8 ppm; skin contained concentrations averaging about 14 ppm. The larger shrew, Blarina brevicauda, exhibited a trend of decreasing DDT levels in its tissues (Fig. 5). Fat, the tissue with the highest residue burden, declined from 600 ppm in June and August 1970 to 150 ppm in 1974. Liver and brain declined from 1970 levels of 15 and 8 ppm to 10 and 9 ppm, respectively, in 1974.

Tissues of the garter snake, Thamnophis sirtalis, did not exhibit consistent trends in DDT levels over time (Fig. 6). Fat contained about 54 ppm in 1970-1972 and declined to 20 ppm in 1973. Liver residues averaged 6.5 ppm in 1971 and 1972, compared to levels near 4 ppm in 1970 and 1973. Muscle increased steadily from 1 ppm in 1969 to 6.3 ppm in 1972, then decreased to 4.8 ppm in 1973.

Estimation of Biomass

Collections of clip plots in 1970 indicated that peak biomass of standing green vegetation occurred in July and September (Table 7). As the differences were relatively small among the months of July, August and September, a combined "midsummer standing crop" of $323 \pm 21 \text{ g/m}^2$ was calculated by averaging the biomass for the three months. This value was converted to 1561 g/m^2 living vegetation by assuming an average moisture content of 80%. Mean earthworm biomass did not vary significantly between May 1971 and November 1973. All the data were therefore combined for a mean ($\pm 1\text{SE}$) live weight of

Table 7. Biomass of vegetation from clip-plots collected in the treated area during 1970. Data given as mean (\pm SE) dry weight in gm/m².

Plant Material	Collection Date					
	6/17	7/18	8/15	9/15	10/15	11/16
Roots	90.4 \pm 13.6	71.5 \pm 10.6	70.4 \pm 9.6	92.0 \pm 12.4	98.8 \pm 7.6	119.6 \pm 10.0
Detritus	40.4 \pm 8.8	34.1 \pm 4.6	64.4 \pm 19.2	20.4 \pm 3.2	23.6 \pm 5.6	12.8 \pm 1.6
Litter	463.2 \pm 53.2	432.0 \pm 37.1	505.2 \pm 27.6	589.6 \pm 61.2	638.8 \pm 53.2	771.2 \pm 58.4
<u>Agropyron repens</u>	67.6 \pm 30.0	78.6 \pm 26.9	146.0 \pm 34.0	36.0 \pm 13.2	24.7 \pm 10.0	33.2 \pm 11.1
<u>Poa sp.</u>	70.4 \pm 20.0	61.9 \pm 12.8	33.2 \pm 8.4	54.8 \pm 15.2	72.4 \pm 14.4	40.0 \pm 7.2
<u>Pastinaca sativa</u>	71.2 \pm 25.6	71.6 \pm 28.0	0.4 \pm 0.2	4.0 \pm 0.2	2.4 \pm 0.4	1.1 \pm 0.4
<u>Dactylis glomerata</u>	41.2 \pm 26.8	29.8 \pm 3.1	64.8 \pm 39.2	94.8 \pm 41.2	59.6 \pm 37.6	113.6 \pm 43.6
<u>Daucus carota</u>	0.4 \pm 0.3	4.2 \pm 1.4	2.6 \pm 1.6	16.0 \pm 6.8	8.8 \pm 2.8	1.9 \pm 1.0
<u>Achillea millefolium</u>	9.8 \pm 6.6	1.4 \pm 1.1	1.2 \pm 0.9	7.6 \pm 5.6	0.0	0.3 \pm 0.2
<u>Solidago juncea</u>	7.7 \pm 1.3	71.9 \pm 44.0	16.2 \pm 16.2	7.0 \pm 5.8	7.2 \pm 4.9	0.0
<u>Aster ericoides</u>	0	6.5 \pm 3.3	9.9 \pm 6.0	49.7 \pm 22.7	44.0 \pm 16.8	1.6 \pm 0.7
Other species	6.9 \pm 3.1	24.3 \pm 9.2	26.2 \pm 6.6	56.0 \pm 19.9	6.7 \pm 2.3	4.2 \pm 2.3
Total Green Biomass	274.8 \pm 33.2	343.4 \pm 39.4	300.0 \pm 33.2	326.0 \pm 35.6	226.0 \pm 27.2	195.9 \pm 38.5

Table 8. Numbers and biomass of Microtus pennsylvanicus determined by livetrapping on alternate days in 1970, and daily in 1971 and 1973.

Period	Trap nights*	Biomass in grams					
		Adults	N	Sub-Adults**	N	Total	N
<u>1970</u>							
July 3-31	2780	1755.8	57	443.4	29	2199.2	86
August 3-31	2600	1518.0	48	590.3	31	2108.3	79
September 2-30	2600	1311.5	44	1103.8	64	2415.3	108
October 2-30	2280	1065.5	29	1006.1	50	2071.6	79
November 2-30	1800	472.2	15	712.3	35	1184.5	50
<u>1971</u>							
June 2, 10 & 30	280	0.0	0	0.0	0	0.0	0
July 14-17	400	0.0	0	0.0	0	0.0	0
September 10-16	800	0.0	0	0.0	0	0.0	0
October 7-13	700	0.0	0	0.0	0	0.0	0
<u>1973</u>							
July 21-31	1100	4267.3	116	1023.9	69	5291.2	185
August 23-29	600	4110.2	109	1605.1	93	5715.3	202
September 23-28	600	3344.6	81	1701.1	100	5045.7	181
October 21-26	600	1771.5	50	1572.2	73	3343.7	123
November 18-23	600	743.0	23	910.8	42	1653.8	65

* One trap night equals one trap set for one night.

** Subadults were small nonbreeding voles, ranging from 9 to 24 g in body weight at first capture.

Table 9. Numbers and biomass of Blarina brevicauda determined by livetrapping on alternate days in 1970, and daily in 1971 and 1973.

Period	Trap nights*	Biomass in grams					
		Adults	N	Sub- adults**	N	Total	N
<u>1970</u>							
July 3-31	2780	365.4	20	786.7	56	1152.1	76
August 3-31	2600	56.5	3	279.0	18	335.5	21
September 2-30	2600	89.0	5	416.7	25	505.7	30
October 2-30	2280	39.2	2	172.8	12	212.0	14
November 2-30	1800	35.2	2	34.5	2	69.7	4
<u>1971</u>							
June 2, 10 & 30	280	0.0	0	0.0	0	0.0	0
July 14-17	400	0.0	0	0.0	0	0.0	0
September 10-16	800	36.9	2	87.8	6	124.7	8
October 7-13	700	52.8	3	193.3	13	246.1	16
<u>1973</u>							
July 21-31	1100	686.5	35	1683.3	111	2369.8	146
August 23-29	600	415.4	21	1248.3	78	1663.7	99
September 23-28	600	350.2	17	1379.9	77	1730.1	94
October 21-26	600	238.0	13	1235.0	78	1473.0	91
November 18-23	600	134.7	8	1119.6	71	1254.3	77

* One trap night equals one trap set for one night.

** Subadults were small, nonbreeding shrews ranging from 11 to 18 g in body weight at first capture.

Table 10. Numbers and biomass of Sorex cinereus determined by livetrapping on alternate days in 1970, and daily in 1971 and 1973.

Biomass in grams							
Period	Trap nights*	Adults	N	Sub-adults**	N	Total	N
<u>1970</u>							
July 3-31	2780	74.1	19	89.6	32	163.7	51
August 3-31	2600	42.9	11	84.0	30	126.9	41
September 2-30	2600	3.9	1	28.1	10	32.0	11
October 2-30	2280	7.8	2	131.6	47	139.4	49
November 2-30	1800	0.0	0	44.8	16	44.8	16
<u>1971</u>							
June 2, 10 & 30	280	0.0	0	0.0	0	0.0	0
July 14-17	400	0.0	0	0.0	0	0.0	0
September 10-16	800	15.6	4	64.4	23	80.0	27
October 7-13	700	27.3	7	131.6	47	158.9	54
<u>1973</u>							
July 21-31	1100	23.4	6	5.6	2	29.0	8
August 23-29	600	11.7	3	0.0	0	11.7	3
September 23-28	600	7.8	2	19.6	7	27.4	9
October 21-26	600	0.0	0	31.2	12	31.2	12
November 18-23	600	3.9	1	10.4	4	14.3	4

* One trap night equals one trap set for one night.

** Subadults were small, nonbreeding shrews ranging from 2.1 to 3.1 g in body weight at first capture.

$47.5 \pm 3.3 \text{ g/m}^2$, representing 159 collections. Isopods attained maximum numbers in October and November; hence, the peak biomass for these two months was $2.5 \text{ g live weight per m}^2$ (N=20). As slugs did not vary significantly in biomass from July through November, the mean standing crop in live weight was 0.81 ± 0.17 (N=50). The cricket, Nemobius allardi, attained peak living biomass of $1.43 \pm 0.46 \text{ g/m}^2$ in September (N=10). Firefly larvae did not exhibit a temporal trend in numbers; therefore the biomass for July-November was 0.54 ± 0.14 (N=50) in live weight.

Peak populations of Microtus occurred in 1970 and 1973; none were live-trapped in 1971 (Table 8). Snap-trapping for residue analysis indicated that numbers were increasing in the late summer and autumn of 1972. The peak biomass values of September 1970 and August 1973 were combined and corrected for 15% evasion of traps (Hilborn et al. 1976) to provide a peak biomass of 4782.7 g for the 4.05 ha enclosure. Vole numbers appeared to be cycling in a typical manner, attaining peaks at 3-year intervals (Myers and Krebs 1971). The residual population between peaks was estimated by comparing Blarina and Microtus biomass in 1970 and 1973 (Tables 8 and 9). Peak Microtus biomass was 2.1 times that of Blarina in 1970 and 2.6 that of Blarina in 1973, averaging 2.3. Since Blarina also declined severely in 1971, it is postulated that Microtus biomass was 2.3 times the peak Blarina biomass in 1971, or 566.0 g. This biomass represents only about 2.3 voles per acre (5.7/ha).

The data of Buckner (1966) indicate annual peaks in shrew numbers (Sorex and Blarina) with relative peak heights varying among habitats. Estimations of peak biomass for Blarina were averaged over the three years (Table 9), giving a value of 1477.6 g corrected for trap evasion.

The trapping results for Sorex cinereus (Table 10) indicated that as the July-August population was removed, a second generation of young shrews produced during August and September entered the trappable population in October (Forsyth 1976). The midsummer peak was estimated to be 114 shrews, 37.3% of which were adult. The 1971 population peaked in October and was estimated to be 82 shrews, 14.8% adult. The 1973 population was estimated to be 26 individuals in the September-October peak, 28.6% of which were adult. Using the mean subadult and adult weights of 2.8 and 3.9 g, respectively (Forsyth 1976), peak biomass was estimated to be 365.9, 242.9 and 81.0 g for the three years, with an average value of 229.9 g.

DISCUSSION

Residue data for soil (Table 2) indicate that 77% of the amount deposited in granules in 1969 remained in the soil in 1971 and declined to 52.7% in 1974. The decline between 1971 and 1974 may not have been real, however, as the change was not statistically significant and soil levels appeared to increase in 1972 and 1973.

Grau (1978) reported that volatilization of DDT ended within five months post-application. The fact that volatilization was occurring in the present study in the second and third years post-application (Table 5) may be explained by the formation of volatile metabolites, especially DDE.

The phenomenon of residues increasing in plants, Microtus, Sorex, isopods, crickets and carabid beetles may also have been a result of formation of DDE with time. As the metabolite formed, it may have percolated deeper into the plant root zone for absorption, or it may have volatilized in increasing amounts and adhered to plant surfaces. Dense grass cover would have trapped more DDT vapor, hence the higher residues in grasses, or, grass roots being more extensive than those of herbs, may have absorbed greater quantities. Animals showing increasing residue loads with time probably acquired them directly from plants or from herbivorous prey. Such predators might include Sorex, Nemobius crickets and Abacidus beetles.

Residues were evidently becoming less available to earthworms, slugs, firefly larvae and Blarina with passage of time. Possibly the invertebrates of this group obtained DDT directly from soil in which the chemical was becoming progressively more bound to organic matter. In feeding to a large extent on earthworms (Whitaker and Mumford 1972) and other soil invertebrates, Blarina would be expected to exhibit a similar pattern of declining residues.

The DDT content of plants, earthworms, slugs and isopods was plotted against soil levels as indicated by granule deposition

(Fig. 7) primarily to obtain estimates of residues for plants and earthworms that would be representative of the study area as a whole, to be used in calculating residue loads in biotic compartments. The median Cl-36 activity of the study area was 2230 dmp (N=430) determined from the granule deposition data of Bandy and Peterle (1972:219). The expected residues in plants and earthworms at this level of soil activity are given in Table 11. It is also evident from Fig. 7 that, whereas plants and earthworms remain at fixed sites so that tissue residues correspond linearly with soil residues, isopods are relatively mobile, resulting in a more curvilinear relationship. Isopods wander to the extent that even specimens collected at sites with expected soil levels close to zero have residues in the 5-7 ppm range.

The annual biomass values calculated earlier have been combined with mean annual (averaged over the entire growing season) residue data for the corresponding species in Table 11. The largest compartment was vegetation, which increased in residue load from 101.1 gm of DDT in 1969 to 396.2 g in 1974, as residues increased. Earthworms were the second largest compartment, declining from a maximum of 105.9 g of DDT in 1969 to 5.6 g in 1974. All the invertebrates exceeded the small mammals in residue load. Total DDT bound in the organisms listed in Table 11 was 208.7 g in 1969 and had increased to 403.5 g by 1974, primarily due to increasing residues in the vegetation.

Soil residue loads in 1971 and 1974 were calculated by graphing ppm in the top 6 cm of 10 sites as a function of Cl-36 in clay granules, similar to the plots shown in Fig. 7 for biota. The

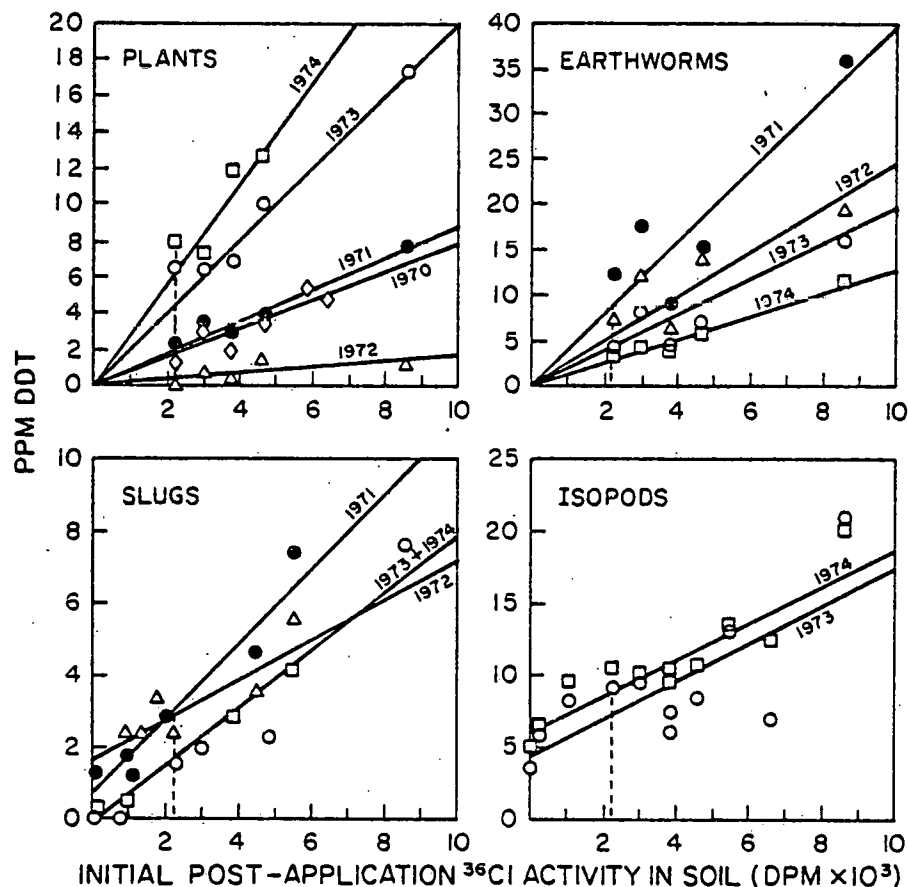


Figure 7. Relationship between DDT residues in plant and invertebrates and expected residues in soil at site of collection. Expected soil residues were determined by analysis of DDT-treated granules collected in petri-dish samples during application. Data points represent annual means. The lines were fitted by linear regression analysis. Vertical dashed lines indicate residues in organisms representative of the median level of Cl-36 activity in soil (2230 dpm).

Table 11. Compartmentalization of DDT residues in major biota of the study area. Annual mean residues refer to whole organism for all species.

Species	Peak Biomass (g/m ²)	Total Biomass (g)	Biomass (g) x Annual Mean Residues (µg/g)					
			1969	1970	1971	1972	1973	1974
Earthworm	47.54	1,924,704	55*	17.63	8.77	5.60	4.04	2.90
	Total DDT	(µgm x 10 ⁶) =	105.9	33.9	16.9	10.8	7.8	5.6
Isopods	2.50	101,215	0.0	N.S.C.	2.51	4.59	8.95	10.80
	Total DDT	(µg x 10 ⁶) =	0.0	--	0.25	0.46	0.91	1.09
Slugs	0.81	32,793	28.93	9.33	3.04	2.96	1.66	1.66
	Total DDT	(µg x 10 ⁶) =	0.95	0.31	0.10	0.10	0.05	0.05
Nemobius	1.43	57,895	2.58	2.18	2.01	4.7*	7.39	7.4*
	Total DDT	(µg x 10 ⁶) =	0.15	0.13	0.12	0.3	0.43	0.4
Firefly	0.54	21,862	25*	12.09	7.8*	5.7*	4.52	4.75
	Total DDT	(µg x 10 ⁶) =	0.5	0.26	0.2	0.1	0.10	0.10
Sorex	0.006	230	3.2	4.0	7.3	9.4	9.2	9.6
	Total DDT	(µg) =	736	920	1679	2162	2116	2208
Blarina	0.036	1,478	71.2	32.2	28.4	24.3	21.6	16.7
	Total DDT	(µg x 10 ³) =	105.2	47.6	42.0	36.0	31.9	24.7
Microtus	0.118	4,783		2.73			5.88	
	Total DDT	(µg x 10 ³) =		13.1			28.1	
Microtus	0.016	566	0.17		3.7*	4.66		6.81
	Total DDT	(µg) =	96.2		2094	2638		3854
Vegetation	1561	63,194,597	1.60	1.73	1.98	0.42	4.54	6.27
	Total DDT	(µg x 10 ⁶) =	101.1	109.3	125.1	26.5	286.9	396.2
Annual	Total DDT	(g) =	208.7	144.0	142.7	38.3	296.3	403.5

* Estimated from logarithmic plots of available data

N.S.C. - No samples collected

resulting plots were linear through the range 0-5000 dpm Cl-36, and the soil residues predicted by substituting the median value for distribution of clay granules in the study area (2230 dpm) into the regression equations were 0.88 ppm for 1971 and 0.66 ppm for 1974. The surface area of a soil core was 20.268 cm^2 ; therefore, there were 493.4 cores per square meter. The residue concentrations represent 70.4 and 52.8 μgm per 6 cm soil core in 1971 and 1974, respectively, or 34,734.6 μgm and 26,050.9 μgm per square meter. Thus, in the 4.05 ha plot, the DDT remaining in the top 6 cm of soil was 1406.3 g in 1971 and 1054.7 g in 1974. These values were increased by 1.7% to include the DDT below 6 cm, resulting in 1430.6 g in 1971 and 1072.9 g in 1974. Adding the DDT present in biota, the DDT remaining in the ecosystem was 1573.3 g in 1971 and 1476.4 in 1974. This decrease in three years was only 6.2%, compared to a decrease of 25% in the soil during the same interval. Increasing residues in plants were compensating in part for loss from the soil. These calculations assume a real loss of DDT from soil, although the difference between mean residues of soil from 10 sites was not statistically significant between 1971 and 1974 (Table 2). Confirmation of volatilization occurring in 1972 (Table 5) suggests nonetheless that the soil was losing residues. Using the data of Table 2 and assuming a linear decline of soil residues with time, DDT would be expected to disappear from the ecosystem in 12 years post-application, that is, by June 1980. This rate of loss is not likely to occur,

however, as indicated by studies reported in the literature. Nash and Woolson (1967) calculated that 10.5 years would be required for soil residues to decline to half of the original amount, and Dimond et al. (1970) reported no detectable decline in DDT content of forest soil nine years post-application.

Our results indicate that in 1974, five years post-application, the soil contained 23.3% of the original quantity of DDT (4634 g) that was applied to the enclosure and the biota contained 8.7%.

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PART II

TRANSLOCATION AND ACCUMULATION
OF ^{14}C -DDT IN AN EXPERIMENTAL OLD-FIELD PLOT

A Thesis

Presented in Partial Fulfillment of the Requirements
for the Degree Master of Science

by

Brenda Lynn Grau, B.S.

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Advisor

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INTRODUCTION

Literature Review

The compound, 2,2-bis-(p-chlorophenyl)-1,1,1-trichloroethane (DDT), was first synthesized by a German chemist, Zeidler, in 1874. Its insecticidal properties were discovered in 1939 by Paul Muller of the Geigy Company of Switzerland. DDT was extremely successful in its use as a synthetic insecticide because of a combination of properties, including ease and low cost of production due to its simple structure, prolonged persistence, non-specificity as a poison, and relatively low mammalian toxicity (Metcalf, 1955:127). The persistence of DDT can be attributed to its resistance to breakdown by sunlight and weathering. The half-life of DDT in the environment has been estimated to range from 7.75 months to 35 years, with the most common estimate being 10 years (Fleck 1944; Terriere and Ingalsbe 1953; Lichtenstein and Schulz 1959a; Woodwell and Martin 1964; Nash and Woolson 1967; Woodwell 1967; Dimond et al. 1970; Woodwell et al. 1971; Menzie 1972). Lichtenstein and Schulz (1959a) demonstrated that the environmental half-life of DDT was directly related to the application concentration. Persistence is enhanced by the compound's extremely low vapor pressure

(1.5×10^{-7} mm Hg at 20°C.) (Balson 1947), its relative insolubility in water (1.2 ppb at 25°C.), and its apolarity (Bowman et al. 1960). Just as DDT is highly insoluble in water, it is highly soluble in lipids. It is this characteristic that is responsible for the accumulation of DDT in the fatty tissues of living organisms and consequently, its potential as an environmental hazard. The hazard is further compounded by the non-selectivity of DDT as a poison, the very quality that makes it so useful as an insecticide. Due to the environmental hazards of DDT and its possible carcinogenic effects on man, the chemical was banned from use in the United States in 1972 (U.S. Environmental Protection Agency, 1975).

DDT and other organochlorine insecticides are present in the atmosphere world-wide, even in places remote from the nearest point of pesticide application (Antommaria et al. 1965; Tatton and Ruzicka 1967; Risebrough et al. 1968; Woodwell et al. 1971; Edwards 1973). DDT enters the atmosphere via direct drift during the application process (Abbott et al. 1966; Pionke and Chesters 1973; Spencer 1975), volatilization from treated soils (Abbott et al. 1966; Pionke and Chesters 1973; Gerakis and Sficas 1974; Spencer 1975), possible codistillation with water (Acree et al. 1963; Bowman et al. 1965; Risebrough et al. 1968; Gerakis and Sficas 1974), and with wind blown dust particles from soil surfaces containing DDT residues (Cohen

and Pinkerton 1966; Pionke and Chesters 1973; Spencer 1975).

Most of the DDT in the atmosphere is adsorbed to particulate matter (Abbott et al. 1965; Stanley et al. 1971; Woodwell et al. 1971; Södergren 1973) and may be carried many miles from its point of origin by the wind and air currents (Antommaria et al. 1965; Cohen and Pinkerton 1966; Tatton and Ruzicka 1967; Risebrough et al. 1968; Tarrant and Tatton 1968; Woodwell et al. 1971; Edwards 1973). Spencer (1975) stated that DDT is probably lost from the atmosphere by rainout, fallout of particulate matter, or photochemical degradation into other compounds. Woodwell et al. (1971) reported that rainfall is the dominant removal mechanism.

Just as DDT is found throughout the world's atmosphere, it is also present in global water systems. DDT is deposited world-wide as it is removed from the atmosphere by rain or snow. Residues have been reported in places as remote from the use of DDT as Antarctica (George and Frear 1966; Peterle 1969). Once deposited on the earth's surface by rain or snow, DDT is again removed and transported to other areas, primarily by surface run-off (Chisholm and Koblitsky 1959; Hindin et al. 1966; Sparr et al. 1966; Freed 1970; Pionke and Chesters 1973; Willis and Hamilton 1973; Gerakis and Sficas 1974). The DDT transported by run-off is mainly associated with the particulate matter and not the water itself (Freed 1970; Pionke and Chesters 1973; Södergren 1973; Holden 1975). It is this transport of DDT

in the run-off that is considered to be of major importance in the accumulation of residues in the oceans (Risebrough et al. 1968; Woodwell et al. 1971; Pionke and Chesters 1973). Even though DDT is relatively insoluble in water (Bowman et al. 1960; Metcalf 1971; Woodwell et al. 1971), once into the water systems, it is very persistent (Lichtenstein and Schulz 1961), probably because of its adsorption to particulate matter. Organochlorine insecticides are generally considered to be non-leachable in soils; hence, very little contamination of ground waters occurs (Bowman et al. 1965; Sparr et al. 1966; Guenzi and Beard 1967; Freed 1970; Pionke and Chesters 1973; Willis and Hamilton 1973; Gerakis and Sficas 1974; Achari et al. 1975).

The persistence and movement of DDT in soil are dependent upon several factors. The method of application (surface or soil incorporated) and the kind of formulation (granular, spray, or powder) are important with regard to the immediacy of loss of the pesticide into the atmosphere and run-off. Organochlorine insecticides are lost more rapidly when applied to the soil surface than when mixed into the soil (Chisholm et al. 1950; Lichtenstein and Schulz 1961; Wheatley 1965; Hindin et al. 1966), and granular formulations persist longer than any other kind of formulation (Edwards 1966; 1973). The rate of loss of DDT from a treated soil immediately after application is higher than subsequent rates of loss. The rate of loss is not

linear and slows with time (Lichtenstein and Schulz 1959a; Edwards 1973; Pionke and Chesters 1973). The percentage of DDT lost from treated soils tends to be inversely proportional to the original application rate, whereas the environmental half-life of the compound tends to vary directly with the application rate (Lichtenstein 1957; Lichtenstein and Schulz 1959a; Nash and Woolson 1967; Holden 1975). Soils with a high organic matter and/or clay content retain DDT for much longer periods than do more mineral soils (Lichtenstein and Schulz 1959b; Lichtenstein et al. 1960; Bowman et al. 1965; Edwards 1966; Guenzi and Beard 1970; Peterson et al. 1971; Woodwell et al. 1971; Edwards 1973; Spencer et al. 1974). The rates of volatilization and degradation of the organochlorine insecticides are directly proportional to temperature, either of the soil or the ambient air (Kiigemagi et al. 1958; Chisholm and Koblitsky 1959; Lichtenstein and Schulz 1959a; Farmer et al. 1972; Guenzi and Beard 1976; Willis et al. 1976) and also to relative humidity (Barlow and Hadaway 1956; Harris and Lichtenstein 1961; Johnsen and Starr 1967). DDT is less stable in alkali than in acid soils, thereby persisting longer in soils with a low pH (Edwards 1966; Nash and Woolson 1967; Nash et al. 1973). Soil moisture content affects persistence in that DDT will bind tightly to all available adsorption sites in dry soil. In these soils, loss is minimal and persistence high. In moist soils,

water will compete with DDT for adsorption sites, hence leaving more pesticide available for possible volatilization (Barlow and Hadaway 1956; Lichtenstein et al. 1960; Bowman et al. 1965; Edwards 1966; 1973). Volatilization of DDT from the surface of treated soils is considered to be a major route of loss (Chisholm and Koblitsky 1959; Woodwell et al. 1971; Edwards 1973; Pionke and Chesters 1973; Spencer et al. 1973; Haque and Freed 1974). There is no definitive evidence concerning the process whereby DDT reaches the soil surface for volatilization, but 2 theories have been set forth; "codistillation" of the pesticide with water (Bowman et al. 1959; 1960; 1965; Weidhaas et al. 1960; Acree et al. 1963) and the "wick effect" where a suction gradient created by evaporating water draws the pesticide to the soil's surface (Lichtenstein and Schulz 1961; Hartley 1969, as cited in Spencer and Cliath 1973; Spencer et al. 1969; Igue et al. 1972; Spencer and Cliath 1969; 1970; 1973; Spencer 1975).

Description of Problem and Objectives

In 1969, a study was initiated to trace the accumulation and translocation of chlorine-36 ring-labeled DDT in an intact old-field ecosystem (Bandy 1972; Bandy and Peterle 1972). In that study, the effects and accumulation of the pesticide in old-field flora and fauna were measured. Sufficient measurements of DDT in air, water, and soil for the immediate post application period were not obtained in that study. A computer model of DDT movement in an old-field

ecosystem was the final goal (Forsyth et al. 1974); DDT movement in the air, water, and soil therefore had to be considered. This movement has been recorded for agricultural lands, including orchards (Chisholm et al. 1950; Chisholm and Koblitsky 1959; Ginsburg and Reed 1954; Ware et al. 1970; Stanley et al. 1971; Bailey et al. 1974), forests (Ide 1956; Chisholm and Koblitsky 1959; Woodwell and Martin 1964; Woodwell 1967; Dimond et al. 1970), and aquatic systems (Ide 1956; Hickey et al. 1966; Meeks 1968; Södergren 1973; Bidleman and Olney 1974; Goerlitz and Law 1974; Haque and Freed 1974) but not for old-field ecosystems. The following 3 objectives were therefore set for the present study:

1. To determine vaporization of ^{14}C -labeled-DDT into the air over an old-field experimental plot.
2. To determine the translocation of ^{14}C -labeled-DDT in the surface run-off, ground water, and soil of an old-field experimental plot.
3. To continue sampling of the old-field fauna sampled by Bandy (1972) and Forsyth (1974) to provide another year of field data for inclusion in the computer model.

The first 2 objectives were met, but no attempt was made to include the additional year of faunal data in this study.

METHODS AND MATERIALS

The study was conducted at the Urbana Wildlife Area in Champaign County, Ohio. Bandy (1972:27-35) set up and described the original 4.05-ha (10-acre) study area. For the present study, a 25-m² plot (Fig. 1a) was chosen near the central portion of the larger area so that drainage would be as uni-directional as possible. A trench extending below the water table was dug around the plot on all four sides. The trench was lined with polyethylene sheeting and backfilled. Galvanized metal sheeting (40 cm wide) was installed on all sides of the plot. An aluminum collecting apron was placed at the NE corner of the plot from the surface to a depth of 10 cm. The apron was triangular shaped and narrowed to fit a notch in a 208-l (55-gal) barrel. The barrel, coated on the inside with teflon, had been placed below surface level for the collection of surface run-off water from the plot (Fig. 1b).

Lysimeters (polyvinylchloride pipes with a porous clay cup at one end) for sampling ground water were placed in the soil of the plot so that the bottom tip of the clay cup was at a depth of 20, 40, or 60 cm. Four pipes were placed at each depth providing a total of 12 lysimeters. Each was capped with a rubber stopper that had a section of vacuum tubing attached (Fig. 1c). The lysimeters were located uniformly throughout the plot but the depth at each location was randomly determined.

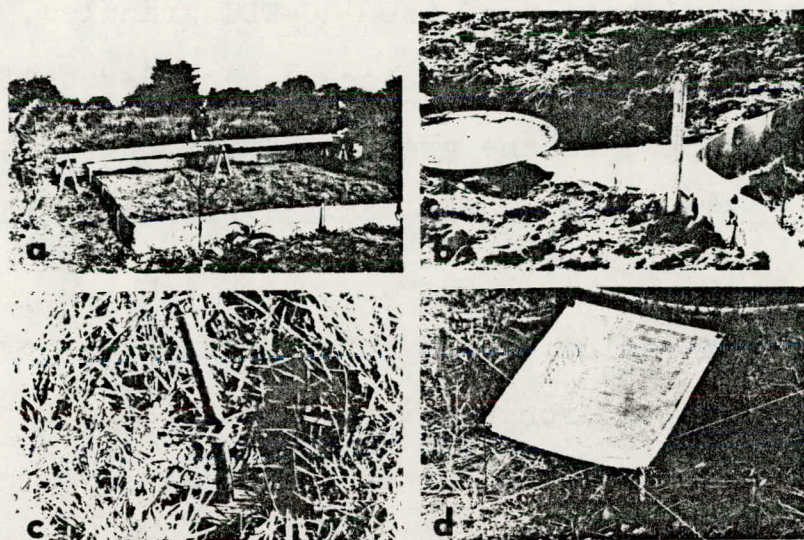


Fig. 1. a. The 25-m² plot immediately prior to the application of ¹⁴C-DDT granules. b. The 208-l drum used for collection of run-off. c. Vacuum tubing and stopper used to seal lysimeters used for collection of ground water. d. Silk screen in place on the plot at the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Prior to application of the ^{14}C -DDT granules, the plot was sub-divided into 25, 1-m^2 blocks. The vegetation on the plot was clipped to the ground. These two procedures facilitated application of the ^{14}C -DDT granules and subsequent sampling.

The uses of radioisotopes for studying the movement of pesticides in the environment were discussed by Peterle (1966). Bandy (1927:22) outlined the reasons for choosing radioisotope labeled DDT for use in our original study. In the present segment of the project, we wanted to be able to differentiate between the labeled DDT applied in 1969 and that applied in 1976. Therefore, a label other than chlorine-36 was needed. Because of the molecular structure of DDT (Fig. 2), we had two choices, either carbon-14 (^{14}C) or tritium (^3H). Carbon-14 was chosen because of its higher transition energy, longer half-life, and greater counting efficiency.

The formulation of the ^{14}C -DDT to be applied to the plot was accomplished by dissolving 2.8 g technical grade DDT in 2.8 g of the emulsifier Triton X-100. Hexane (400 ml) was added to increase the solubility of the DDT. One millicurie (mCi) (12.87×10^{-3} g) of uniformly ring-labeled ^{14}C -DDT in benzene (specific activity 29.7 mCi/mmol; obtained from Amersham/Searle Corporation) was added to the hexane-DDT solution. This solution was then applied to 280 g of attapulgite clay granules (Meeks 1967:31) by

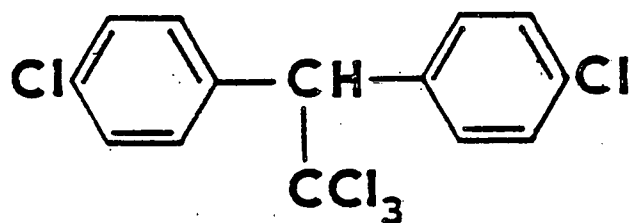


Fig. 2. Structural formula of DDT.

soaking the granules with the ^{14}C -DDT solution and letting the hexane evaporate in a laboratory hood. The reasons granules rather than a spray formulation were used were listed by Bandy (1972:37).

The granules were applied to the plot on 17 June 1976 at a rate of 1 pound of DDT per acre. Each of the 25, 1-m^2 blocks received an even distribution of 11.2 g of ^{14}C -DDT granules, by means of a large salt shaker. A 6-m-long plank that spanned the plot was used during the application process to prevent the investigators from contacting the treated areas.

Field Sampling

Sampling of the plot began on the first day post application. Samples of air, water from surface run-off, ground water, and soil were collected from 18 June to 30 November 1976 and from 29 March to 30 June 1977. Samples of soil, earthworms (Lumbricidae), isopods (Oniscoidea), slugs (Stylommatophora), and shrews (Soricidae) were collected during 1976 from the 4.05-ha study area. These samples were taken to provide an additional year of data on the components of the system about which our information was most complete for the first several years after the original treatment. These samples were not analyzed for this segment of the project.

Air samples were collected via two methods: silk screens impregnated with silicone oil (Fig. 1d) and the Greenburg-Smith impinger system (Bandy 1972:63), which utilized two collection media, ethylene glycol and Chromosorb 102 (an adsorbent added to the impinger system). Silk screens were used for collecting air samples as a modification of Södergren's (1974) technique. Each screen (841 cm^2) was placed on a stainless steel frame, coated with $1.6 \pm 0.1 \text{ g}$ silicone oil (Dow Corning 704), and set 20 cm above the ground at a 30° angle above the horizontal. Sixteen screens were distributed uniformly on the experimental plot. An additional 16 screens were set out as controls (background) about 100 m SW of the 25-m^2 plot. Two control and 2 experimental screens were collected at the end of each month and stored in glass jars with foil-lined caps pending laboratory analysis. Two trains of the Greenburg-Smith impinger system (obtained from the U.S. Public Health Service) were used for air sample collection (Bandy 1972:62). They were connected sequentially and each was charged with 100 ml ethylene glycol. The machine was run 8 h per day for 3 days during the summer of 1976 for a total of 24 h per sample. From September through November 1976 and in all 1977 samples, the machine was run for 3.5 h per day for 7 days for a total of 24.5 h per sample. Samples were taken during the hottest part of the day. The flow-meter of the machine

was set so that air was collected at a rate of 2.83×10^{-2} m³ per minute. Thus, the 24-h samples represented an air volume of 40.75 m³ and the 24.5-h samples represented a volume of 41.60 m³. Air was collected at ground level by laying perforated polyethylene hoses over the plot and connecting them to the impinger system. The hoses were moved to different sections of the plot after each sample was collected and the impingers were recharged. Control (background) samples were taken by laying the hoses outside the 25-m² area for the sampling period. Chromosorb 102 (Thomas and Seiber 1974) was added to the impinger system just below the intake filter. Five grams were used in each train of the impinger. A plug of glass wool prevented the adsorbent from falling out of place and contaminating the ethylene glycol below. Both the ethylene glycol and the Chromosorb 102 were placed in glass jars with foil-lined caps upon collection. Each section of the impinger that had contained a collection medium was rinsed 3 times with acetone, which was then added to the collection jars (about 25 ml for ethylene glycol and 10 ml for Chromosorb 102). Each train was thoroughly washed and rinsed with acetone and hexane before it was recharged.

Surface run-off water was collected in a 208-l (55-gal) teflon-coated drum at the NE corner of the plot. To aid in collection of the run-off, a large trash can, lined with a plastic bag, was placed in the drum. This trash can could be lifted out of the drum for sample collection. The plastic

bag was replaced after each collection. Collection dates were determined by the rainfall pattern at the study area. Background samples were taken periodically from a 0.17-ha pond located approximately 100 m NW of the 4.05-ha enclosed area. The samples were stored in glass jars with foil-lined caps pending laboratory analysis.

Collection of ground water samples was also determined by rainfall. Any ground water in the lysimeters was forced out using a hand pump. The sample was pumped into a glass jar capped with a foil-lined lid. After the collection, each lysimeter was evacuated with a hand vacuum pump and sealed until the next collection date. Control (background) samples were taken by pouring 100 ml of distilled water into the evacuated lysimeters and letting them sit overnight. The distilled water was recovered from the pipes the next morning.

Soil samples were collected from 2 of the 25, 1-m² blocks each week. The order in which the blocks were sampled was randomly determined. Each soil sample consisted of 3 cores, each 2 cm in diameter and 6 cm in length. Each 6-cm core was divided into the top 3 cm (including the litter layer) and the next 3 cm. The tops and bottoms of each core were individually wrapped in aluminum foil. The 6 foil-wrapped packages for each sample were frozen pending laboratory analysis. Control (background) samples were collected from an area approximately 100 m N of the

4.05-ha enclosed area and in the same manner as experimental samples.

An attempt was made to collect weather data at the plot, but due to technical difficulties, the collection of these data was inconsistent. Rain and temperature data were obtained instead from records kept by the Urbana Sewage Treatment Plant (approximately 9.6 km SW of the study area).

Laboratory Analysis

Liquid Scintillation Spectrometry

A Packard Tri-Carb Scintillation Spectrometer Model 3003 equipped with Model 574 Automatic Controls and Automatic External Standardization was used to count all samples collected for this study. No distinction between the isomers of DDT and its metabolites could be made with this counting technique. Therefore, only labeled DDT and metabolites were measured (Dindal 1967:26). Scintillation grade glass vials with foil-lined caps were used throughout the counting procedure. The scintillation cocktail was prepared from the formula used by Bandy (1972:46). Quench corrections for ^{14}C were determined by the channels ratio method (Herberg 1965) and the external standardization method (Hendee 1973:211).

Sample Preparation

Air samples collected by silk screens were prepared for counting with a modification of Södergren's (1972) method. Hexane extracts were cleaned with H_2SO_4 by him to allow the use of gas chromatography. The H_2SO_4 was omitted from the air samples because it is a strong quenching agent (Hendee 1973:206). Triton X-100 and cocktail were added to the concentrated extract.

Air samples collected with the Greenburg-Smith impinger system were divided into 2 components for sample preparation: ethylene glycol and Chromosorb 102. The ethylene glycol fraction was prepared with a modification of Bandy's (1972:64) method. The procedure followed was:

1. The ethylene glycol was filtered into a 1-l separatory funnel. The collection bottle was rinsed 3 times with 5 ml acetone which was then filtered. The filter was then rinsed with 5-10 ml acetone. All acetone rinses were added to the filtered ethylene glycol.

2. Hexane (200 ml) was added to the ethylene glycol. The separatory funnel was shaken for 1 minute and then allowed to stand for 1 hour.

3. After an hour, the hexane wash was removed to a clean container and the ethylene glycol was returned to the separatory funnel. Steps 2 and 3 were repeated twice for a total of 3 washes and 600 ml hexane extract.

4. The hexane extract was dried by pouring it through a 20-cm column of anhydrous granular Na_2SO_4 . This step was then repeated. Each Na_2SO_4 column was rinsed with 2, 50-ml aliquots of hexane. The resultant 200 ml of hexane were added to the 600 ml of extract for a total of 800 ml of hexane.

5. The hexane was allowed to evaporate, either with a flash evaporator or in a laboratory hood, to approximately 20 ml. Then, a gentle air stream was applied to speed evaporation to produce 2-3 ml. This quantity was transferred to a scintillation vial and the evaporation flask was rinsed with 3, 2-ml portions of hexane. These rinses were added to the vial.

6. The 8-11 ml of hexane were then further evaporated to 3 ml with a gentle air stream.

7. Triton X-100 (2 ml) and 15 ml scintillation cocktail were added to the hexane as final preparation for liquid scintillation counting.

The Chromosorb 102 fraction was tied into filter paper packets and extracted in a 1:1 mixture of acetone and hexane for 1 hour with a soxhlet apparatus (Thomas and Seiber 1974). The solvent mixture was then evaporated to 5-10 ml and transferred to a scintillation vial. The evaporation flask was rinsed with 5 ml hexane, which was added to the vial. The resulting extract was then further evaporated and prepared for counting as were the ethylene

glycol samples.

The run-off and ground water samples were prepared for liquid scintillation counting by the method outlined by Bandy (1972:67-68). The run-off water was divided into 2 portions. One portion was analyzed for DDT associated with the particulate matter and the other was analyzed for DDT associated with both the particulate matter and water. Ground water samples were prepared only for analysis of DDT associated with the particulate matter and water.

Soil samples were prepared with a modification of Forsyth's (1974) method. The major modifications of this method included wetting the soil prior to extraction and using a 1:1 mixture of acetone and hexane as the extraction solvents. Johnsen and Starr (1970) have reported that these changes greatly improved extraction efficiency. The procedure followed was:

1. Frozen samples were placed in a laboratory hood (the 3 top 3-cm sections and 3 bottom 3-cm sections from each sample were placed in 2 separate dishes) and allowed to thaw and dry.

2. The dried samples were ground with a mortar and pestle. Any roots or detritus were chopped to 0.2-1.0-cm size with scissors.

3. Forty grams each of the top and bottom dried and ground samples were placed in separate 125-ml

polyethylene bottles. If there was not 40 g of soil available, the largest quantity present was used.

4. Distilled water (20 ml) was then added to the bottle to thoroughly wet the soil sample. Equal amounts (20 ml) of both hexane and acetone were then added to the bottle.

5. The bottles of soil samples were shaken for 4 hours in a commercial paint shaker. After shaking, the samples were allowed to stand until the soil had settled. The acetone and hexane mixture was then decanted.

6. The decanted solvent mixture was filtered into an evaporating flask. The soil sample was then washed with 3 separate 20-ml aliquots of hexane which were decanted and filtered. The filtered washes were then added to the acetone-hexane mixture for a total of 100 ml of solvent extract.

7. The extract was dried by pouring it through a 10-cm column of anhydrous granular Na_2SO_4 . This step was then repeated. Each Na_2SO_4 column was rinsed with 10 ml of hexane. The resulting 20 ml of hexane were added to the 100 ml of solvent extract for a total of 120 ml of the acetone-hexane mixture.

8. The extract was evaporated and prepared for counting as were the air samples.

Four soil samples were extracted a second and third time for the determination of extraction efficiency. When

disintegrations per minute (dpm) were plotted as cumulative dpm extracted (abscissa) versus dpm per extraction (ordinate), a linear relationship resulted. The intercept of the line with the abscissa represented the dpm of ^{14}C -DDT present in the soil before extraction and provided the basis for estimating the percent efficiency of extraction (recovery of the ^{14}C -DDT).

After all samples were prepared, they were counted at the optimum gain of each sample type (Table 1). Optimum gains were determined by the method outlined by the Packard Instrument Company (1971:4.4-4.5).

Table 1. Optimum gain settings for samples from the Urbana study area, Champaign County, Ohio, 1976 and 1977. Gain settings expressed as percent of the total machine voltage (Packard Tri-Carb Scintillation Spectrometer Model 3003).

Sample Type	Optimum Gain %
Screens	13.6
Ethylene Glycol	5.0
Chromosorb 102	5.5
Run-Off	
Whole Run-Off	16.0
Particulate Matter	25.2
Ground Water	7.0
Soil	44.0

RESULTS AND DISCUSSION

Air Samples - Screens

Silk screens were analyzed for ^{14}C -DDT residues and the pesticide levels were recorded as ng/m^2 of screen. Carbon-14 DDT residues in screens collected from the study area (Fig. 3) were significantly ($P < 0.01$, two-tailed t-test) higher than those residues in control (background) screens for both 1976 and 1976 and 1977 combined. Residues were analyzed for the 3-month periods of June to August and September to November, 1976. In both periods, experimental screens contained significantly ($P < 0.01$, two-tailed t-test) higher residues than did controls (Table 2). The experimental screens from the June to August period contained much higher quantities of DDT than did the experimental screens from the September to November period ($P < 0.01$, two-tailed t-test). No difference ($P > 0.05$) was detected between the 1976 and 1977 residue data from controls nor between the 1976 control and the 1977 experimental screens. Thus, residues in the experimental screens had declined to background levels by the spring of 1977. These results were perhaps an indication that the screens collected in 1977 had accumulated large amounts of ^{14}C -DDT soon after the application but lost those residues as the screens were exposed to the weather over the months.

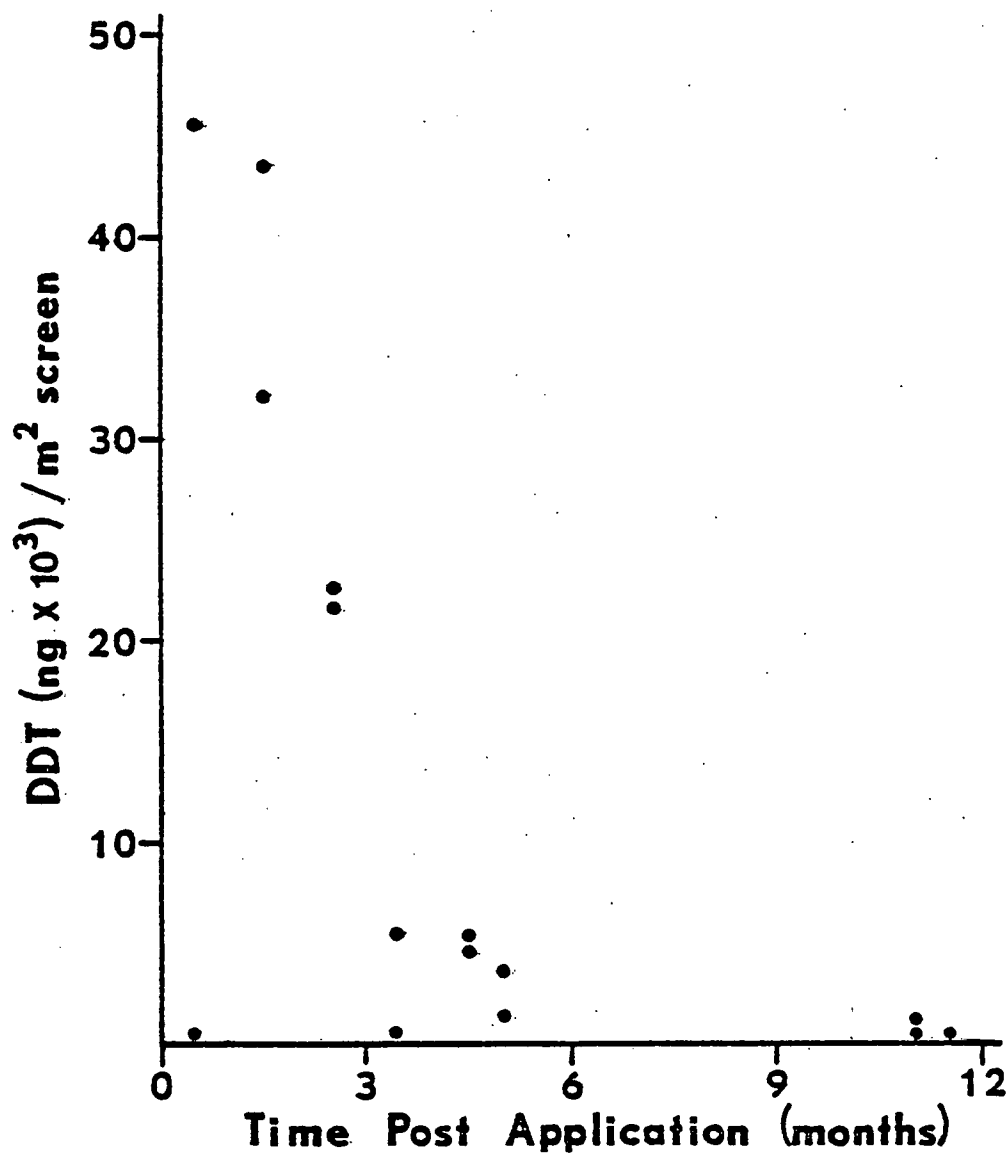


Fig. 3. ¹⁴C-DDT residues (ng/m²) in experimental screens from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Table 2. ^{14}C -DDT residues in silk screens from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Collection Date	DPM		DPM equal to ng DDT / m ² screen	
	Experimental ^a	Control ^a	Experimental ^a	Control ^a
7-01-76 (0.5) ^b	30.38 2954.06	30.11 31.59	469.68 45663.50	464.92 488.70
8-01-76 (1.5)	2070.65 2815.38	32.25 27.95	32007.13 43519.60	498.22 431.63
8-30-76 (2.5)	1412.87 1457.47	39.26 39.27	21839.48 22529.13	607.61 607.61
9-18-76 (3.5)	348.43 26.00	25.78 26.13	5386.44 401.90	398.34 402.28
10-26-76 (4.5)	317.26 333.53	26.53 26.20	4903.69 5155.77	410.23 405.47
11-10-76 (5.0)	75.08 247.27	- -	1160.52 3822.83	- -
11-23-76 (5.5)	- -	27.87 28.00	- -	430.44 432.82
5-11-77 (11.0)	34.44 28.94	27.07 26.42	532.70 447.09	418.55 407.85
6-01-77 (11.5)	32.23 - ^c	25.74 26.93	498.22 - ^c	398.34 416.17

^aTwo experimental and two control screens were collected on each date.

^bNumber of months post application.

^cSample lost.

The amount of ^{14}C -DDT on the experimental screens collected in 1976 was positively correlated ($r = 0.67$) with the amount of time that had passed since application of the pesticide to the study plot. Residues on all experimental screens collected in both years were also positively correlated ($r = 0.60$) with time post application.

The first experimental screen collected had a ^{14}C -DDT residue level equivalent to that of the control samples. This experimental screen was located at the SW corner of the plot, which was the highest part of the 25-m² plot. Possibly the granules that were deposited at this location were dispersed more readily by the rains after application than were granules at more level sections of the plot. The highest levels of DDT in the experimental screens were an order of magnitude greater than the highest levels detected by Södergren (1972) in Sweden.

Air Samples - Ethylene Glycol

Carbon-14 DDT residues in the ethylene glycol samples were recorded as ng/m³ of air. A pre-treatment air sample, collected 2 days before the application of the ^{14}C -DDT granules, contained levels of radioactivity equivalent to those of control (background) samples (Appendix B). Samples collected through 29 days post application contained significantly ($P < 0.05$, two-tailed t-test) higher ^{14}C -DDT

residues than did controls (Appendices A and B). The residues in the ethylene glycol (Fig. 4) were highest in the first sample, collected 3 days post application, and decreased rapidly thereafter. Twenty-nine days after the application of the granules, residues had declined to pre-application and background levels. This rapid reduction of DDT in the air following application agreed with the results of other researchers (Lichtenstein and Schulz 1959a; Edwards 1973; Pionke and Chesters 1973).

Two factors positively correlated with the decline of ^{14}C -DDT residues in ethylene glycol samples were time post application ($r = 0.69$) and the total amount of rain that fell post application ($r = 0.73$). The amount of rain that fell during each collection period was also important in the rate of decline of ^{14}C -DDT residues ($r = 0.70$). In accordance with results obtained by other workers (Chisholm et al. 1950; Lichtenstein and Schulz 1961; Wheatley 1965; Hindin et al. 1966), a rapid loss of the DDT into the atmosphere was obtained possibly because the ^{14}C -DDT was surface applied rather than soil incorporated. Another possibility, that of the water in the soil competing with the DDT for binding sites in the soil, thus making more DDT available for volatilization (Barlow and Hadaway 1956; Lichtenstein et al. 1960; Bowman et al. 1965; Edwards 1966; 1973), had to be considered because of the heavy rains after the application.

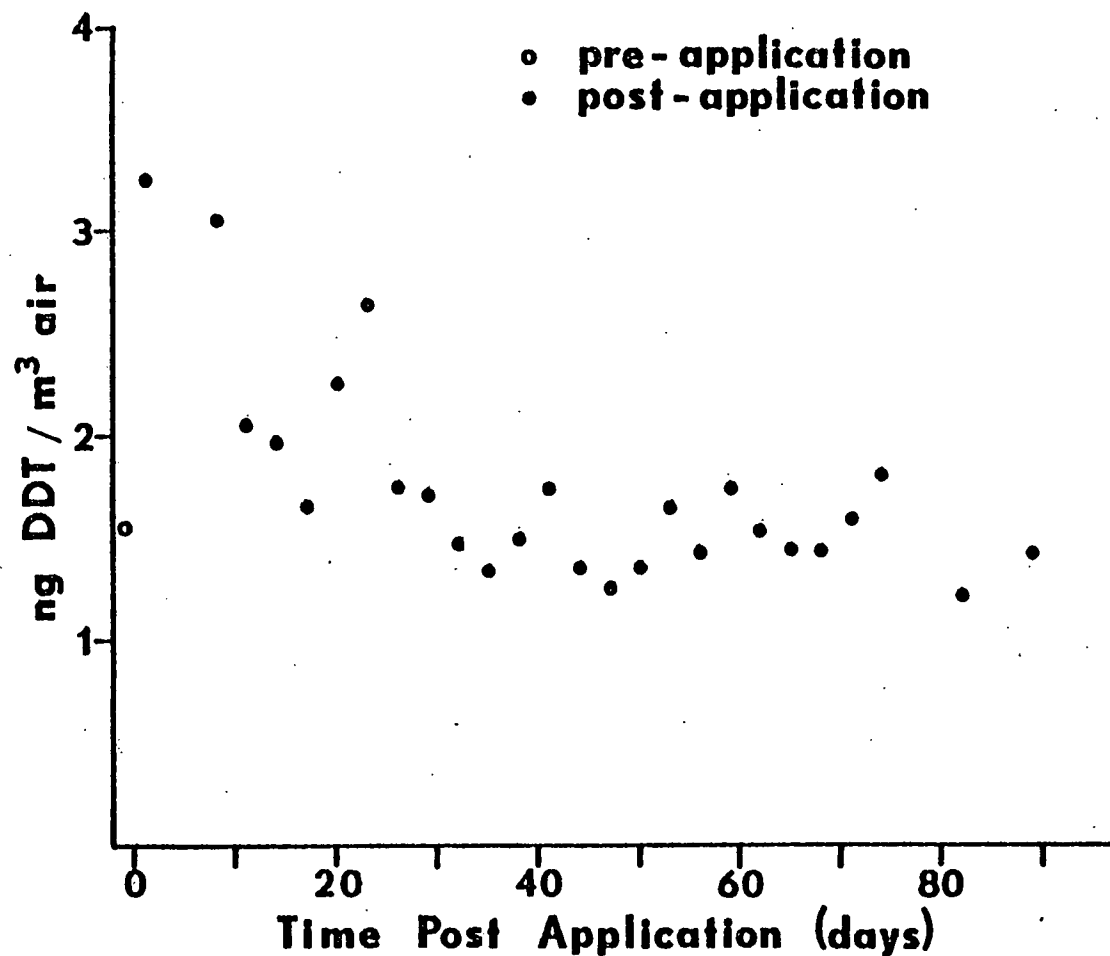


Fig. 4. ^{14}C -DDT residues (ng/m^3) in air (ethylene glycol) samples from the Urbana study area, Champaign County, Ohio, 1976. Each value represents collections from 3 8-h sampling days. The pre-application sample represents the background radiation level.

Temperature ($r = 0.19$) had little effect on the rate of decline of ^{14}C -DDT residues in ethylene glycol samples.

The presence of ^{14}C -DDT residues in air samples at the study area was contrary to the findings of Bandy (1972:145), who found no ^{36}Cl -DDT in air samples with hexylene glycol as the collecting medium. The absence of residues was attributed to the low concentration of insecticide available for sampling and to the high dilution factor involved. One possible reason that Bandy detected no residues in the air was the collection medium; our study used ethylene glycol. One factor not mentioned by Bandy was the height of the air sampler. The intake pipe of the machine was 1.7 m above the ground in Bandy's (1972:64) study, whereas polyethylene tubing placed on the ground and connected to the intake pipe was used in this study.

Carbon- 14 DDT residues were much higher on the screens than in the ethylene glycol. High amounts of ^{14}C -DDT were still in the screens after the ethylene glycol samples had reached background levels. The best explanation for this fact was that the DDT, because it is more soluble in fats and oils than in water (Bowman et al. 1960), was bound in large amounts to the silicone oil used on the screens. Had we been able to use an oil in the Greenburg-Smith impinger system, the residues detected there might have been much higher than they

were with the ethylene glycol.

Air Samples - Chromosorb 102

Addition of Chromosorb 102 to the impinger system began with the eighth sample, which was collected 26 days post application. Upon analysis of the samples, no ^{14}C -DDT residues were found (Appendices C and D). By the time the Chromosorb 102 sampling was initiated, the ethylene glycol had declined almost to background levels. Hence, no difference ($P > 0.05$) was found between background and experimental Chromosorb 102 samples.

Run-Off Samples

Carbon- 14 DDT residues (reported as parts per billion, ppb) were found to be significantly ($P < 0.01$, two-tailed t-test) higher in particulate matter samples than in samples containing both water and particulate matter (whole run-off) (Table 3). These results, inferring association of the DDT with the particulate matter, were compatible with those obtained by other researchers (Freed 1970; Pionke and Chesters 1973; Sodergren 1973; Holden 1975). Residues in particulate matter were highest at 6 days post application and declined rapidly thereafter, reaching background levels by 21 days post application (Fig. 5). Residues in whole run-off samples were highest at 15 days post application. These residues also declined rapidly and reached background levels by 29 days post application.

Table 3. ^{14}C -DDT residues in surface run-off samples from the Urbana study area, Champaign County, Ohio, 1976.

Collection Date	DPM		PPB	
	Particulate Matter ^a	Whole Run-Off ^b	Particulate Matter ^a	Whole Run-Off ^b
6-23-76 (6) ^c	247.22 ^d SD=349.11	408.04 ^d SD=666.57	920 ^d SD=1296	0.73 ^d SD=1.15
7-02-76 (15)	132.00 ^d SD=114.98	264.63 ^d SD=609.68	490 ^d SD=427	0.94 ^d SD=2.37
7-08-76 (21)	44.70	66.11	170	0.20
7-16-76 (29)	42.97 ^d SD=15.40	46.14 ^d SD=12.57	160 ^d SD=57	0.10 ^d SD=0.00
7-23-76 (36)	47.08 ^d SD=15.60	33.82 ^d SD=1.95	170 ^d SD=58	0.10 ^d SD=0.00
7-28-76 (41)	37.76	38.65	140	0.10
8-10-76 (54)	33.92	37.11	130	0.10
9-21-76 (96)	46.78	30.73	170	0.10
9-28-76 (103)	37.50	31.05	140	0.10
10-12-76 (117)	50.06	32.32	190	0.10
10-26-76 (131)	42.39	35.25	160	0.10
11-01-76 (138)	41.71	34.97	150	0.10

^aSample extracted from 50 ml of run-off.

^bSample extracted from 750 ml of run-off.

^cDays post application.

^d \bar{x} DPM and PPB when sample consisted of more than one subsample.

The decline of residue levels in both particulate matter (21 days post application, $r = 0.99$) and whole run-off (29 days post application, $r = 0.79$) samples was highly correlated with time post application. This correlation was probably the result of the surface applied ^{14}C -DDT granules or particles washing off the plot early in the sampling period.

Ground Water Samples

Movement of organochlorine insecticides in soil increases with decreasing molecular weight and with increasing water solubility of the insecticide (McCarty and King 1967, as cited by Caro 1969). Hence, DDT, with the lowest water solubility of any organochlorine insecticide, does not move in soil under conditions that may allow movement of other chlorinated hydrocarbon insecticides (Bowman et al. 1965; Guenzi and Beard 1967). Accordingly, no residues of ^{14}C -DDT were found in the ground water at any of the depths sampled in this study (Appendix E).

Soil Samples

The soil at the study area was a Miami silt loam. Analysis of soil samples from 0-3 cm and 3-6 cm depths was conducted to determine percentages of soil components present that could have affected the retention of DDT residues (Table 4). Due to low amounts of organic matter

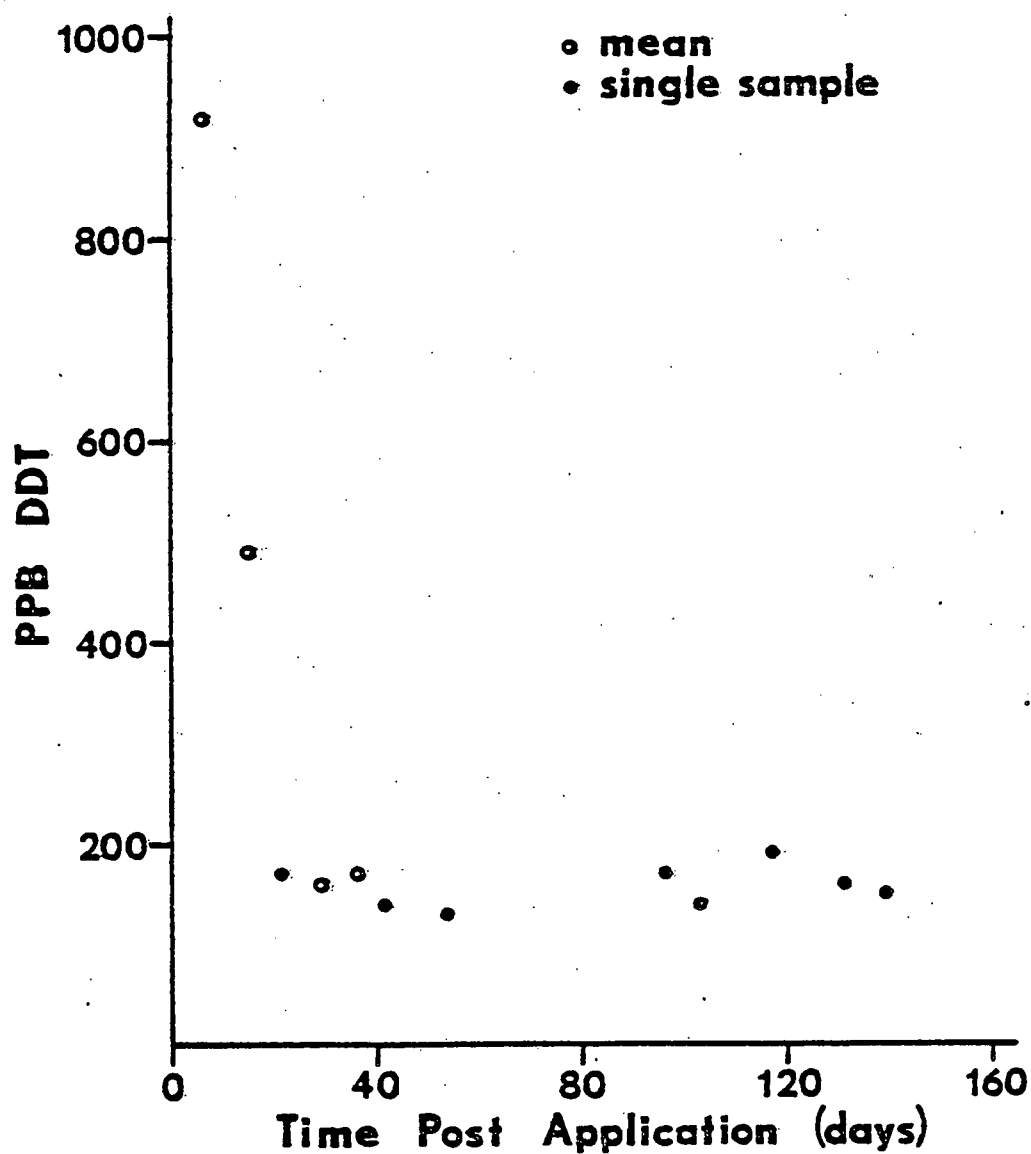


Fig. 5. DDT residues (ppb) in the particulate matter portion of the run-off samples from the Urbana study area, Champaign County, Ohio, 1976. Means were of several subsamples of total run-off. Single samples included all water volume collected.

and clay, soil retention times for DDT were expected to be relatively short (Lichtenstein and Schulz 1959b; Lichtenstein et al. 1960; Bowman et al. 1965; Edwards 1966; 1973; Guenzi and Beard 1970; Peterson et al. 1971; Woodwell et al. 1971; Spencer et al. 1974).

To determine how much of the ^{14}C -DDT present in the soil was actually extracted and counted by LSS, soil extraction efficiency was calculated. Four soil samples, each extracted 3 times, were used to calculate the mean per cent extraction efficiency. Values obtained ranged from 93.22 to 96.86% ($\bar{x} = 95.27$, $\text{SD} = 1.57$). The values obtained were much higher than the 34-76% reported by Forsyth (1974) for ^{36}Cl -DDT. The increased and less variable extraction efficiencies were obtained primarily because of the modifications (wetting the soil prior to extraction and using a 1:1 mixture of acetone and hexane as the extraction solvents) we made in the extraction procedure.

Residues of ^{14}C -DDT (reported as parts per million, ppm) were significantly ($P < 0.01$, two-tailed t-test) higher in the experimental 0-3 cm soil samples than in the experimental 3-6 cm sections for both 1976 and 1977. No statistical difference ($P > 0.05$) was found between the amounts of ^{14}C -DDT present in the soil samples collected in 1976 and those collected in 1977. Residues in the experimental soil samples were greater ($P < 0.01$, two-tailed

Table 4. Characteristics of the soil (Miami Silt Loam) from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Soil Characteristic	0-3 cm	3-6 cm
pH ^a	6.1	6.2
Organic Matter ^a	8.8%	3.9%
Sand ^b	26.4%	19.8%
Silt ^b	66.9%	72.5%
Clay ^b	6.7%	7.7%

^aAnalysis performed by the Soil Testing Service, Ohio Agricultural Research Station, Wooster.

^bAnalysis performed by the Ohio Soil Characterization Laboratory, Department of Agronomy, The Ohio State University, Columbus.

t-test) than those in control (background) samples for both years. Even though amounts of residues did not differ significantly, they declined slightly. The rate of decline in the 0-3 cm experimental samples was positively correlated with the cumulative amount of rain ($r = 0.71$), time post application ($r = 0.68$), and monthly average minimum temperature ($r = 0.68$). Other factors positively correlated with the rate of decline of ^{14}C -DDT residues were the amount of rain that fell each month ($r = 0.65$), monthly average temperature ($r = 0.63$), and monthly average maximum temperature ($r = 0.55$). A slight upward trend in the amount of DDT (mean ppm per month) in the experimental 0-3 cm samples was observed in 1976 (Fig. 6). One-way analysis of variance showed no significant difference ($P > 0.05$) among the means of each collection period. Therefore, DDT residues in the soil did not differ significantly over the time period sampled.

The DDT applied to the study plot dissipated and was absent from air and run-off samples within 30 days. At that time, residue levels in air and run-off were equivalent to background levels. Throughout the 12 months of the study, the levels of ^{14}C -DDT in the soil did not return to control levels (Appendices F, G, and H). Residues of ^{14}C -DDT in the 0-3 cm layer were greater than those in the 3-6 cm layer by 1 to 2 orders of magnitude which were, in turn, greater than background samples by

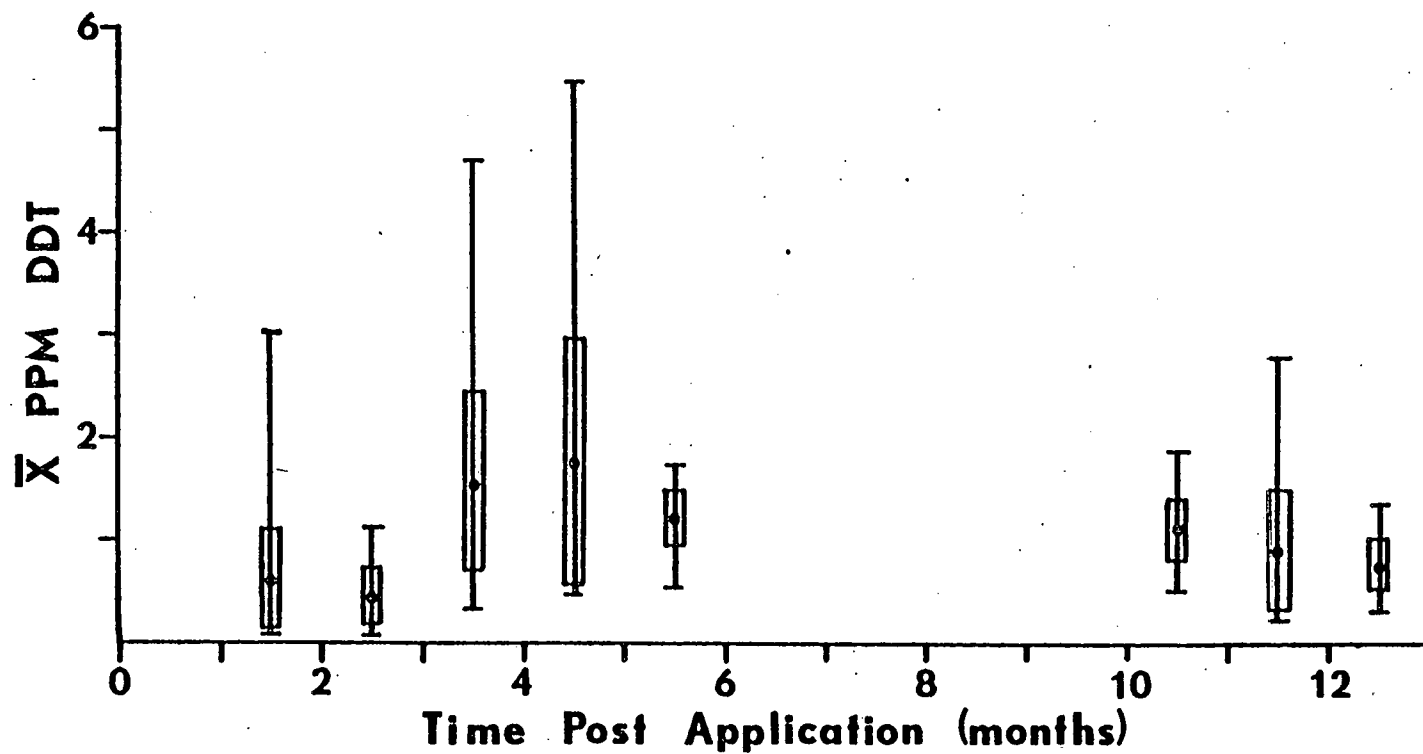


Fig. 6. DDT residues (ppm) in 0-3 cm soil samples from the Urbana study area, Champaign County, Ohio, 1976 and 1977. The symbol gives the range, \pm 2SE, and the mean.

the same margin. Therefore, ^{14}C -DDT levels in the 0-3 cm layer of the experimental plot were greater than those in background samples by 2 to 4 orders of magnitude. Possibly these levels were enhanced because we clipped the vegetation on the plot periodically and allowed the debris to remain on the plot.

Woodwell and Martin (1964) hypothesized the reason for an increase in soil DDT in forests treated only once was due to the addition of DDT contributed by falling leaves. These workers also found that the organic layers of the soil contained more than half of the total DDT and that the increase in DDT from year to year was in these layers. Dimond et al. (1970) concluded that persistent residues were restricted to the upper soil and litter layers in contaminated forests and that there was little decline of residues after 9 years. We cannot compare our results for 1 year with those obtained after 9 years by Dimond et al. (1970) because soil residue levels were not measured for the entire study (begun in 1969) at the Urbana area.

DDT Compartmentalization

The ^{14}C -DDT applied to the plot was eventually distributed in the air, run-off, and soil of the old-field. When this study began, 12.87×10^3 ug (2.22×10^9 dpm) of ^{14}C -DDT and 2.80×10^6 ug of technical DDT (total DDT was

2.81×10^6 ug) had been applied to the plot. On that basis, there were 1.3×10^{-3} ug DDT/dpm in all samples counted by LSS. The amount of the applied DDT in each "compartment" of the old-field plot was determined with the data obtained from each segment of this study (Table 5).

The amount of ^{14}C -DDT detected in the air was more efficiently measured by the silk screens than by the impinger system with ethylene glycol. Assuming all 16 experimental screens had the capacity to hold the maximum amount of ^{14}C -DDT detected in the screens (1954.00 dpm or 3.84 ug DDT), 61.44 ug DDT were held by the screens. The 16 screens covered 5.38% of the area of the plot ($13.46 \times 10^3 \text{ cm}^2$ of $250.00 \times 10^3 \text{ cm}^2$). Had screens covered the entire $250.00 \times 10^3 \text{ cm}^2$ of the plot, the amount of DDT they would have held was determined by extrapolation to be at least 1141.25 ug DDT. The amounts of ^{14}C -DDT in the ethylene glycol samples had declined to background levels by 29 days post application, therefore only those first 29 days were considered in the calculation of the amount of DDT trapped in ethylene glycol. At 29 days post application, 0.83 ug DDT (642.24 dpm) had been detected in 9 ethylene glycol air samples. These 9 samples represented 366.75 m^3 of air collected over a period of 216 hours, an average collection of 3.8×10^{-3} ug DDT/hour. Over the 29-day period, the amount of DDT that could have been collected by the ethylene glycol on

Table 5. Amounts of DDT in each "compartment" of the old-field plot, at the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Compartment	ug DDT	[%] Total DDT
Air		
Screens	1141.25	40.57×10^{-3}
Ethylene Glycol	2.64	0.09×10^{-3}
Run-Off	33.32	1.18×10^{-3}
Soil	2.58×10^6	91.80

the old-field plot had the impinger system been run 24 hours a day for the 29 days was determined; by extrapolation, to be at least 2.64 ug DDT. This estimation assumes that the vaporization of DDT per unit time over the 24 hours was equal to those time periods actually sampled.

For the determination of total DDT in the run-off, the particulate matter and whole run-off data were combined. Because run-off sample volumes were different with each collection, dpm/l of water was determined first for each sample and then converted to ug DDT in the total sample. A total of 118.58 l of run-off was collected and, by extrapolation, at least 33.32 ug DDT were bound in the run-off of the old-field plot.

By the end of this 12-month study, ^{14}C -DDT residues in the soil had not declined to background levels. Therefore, all soil samples collected were used in the determination of the amount of DDT held by the old-field soil. Each soil core (0-3 cm and 3-6 cm) was 2 cm in diameter and contained 3.14 cm^2 of soil. The major portion (94.68%) of the residues detected were in the 0-3 cm soil sections and the ^{14}C -DDT had been applied only to that part of the soil, so only these portions of the samples were used in calculating total soil DDT. The 74 0-3 cm soil samples, with a total of 232.36 cm^2 of soil, contained 2.40×10^3 ug DDT, an average of 10.31 ug DDT/ cm^2 of soil. Because there were $250.00 \times 10^3 \text{ cm}^2$ on the

surface of the plot for the application of the ^{14}C -DDT, by extrapolation, at least 2.58×10^6 ug DDT were present in the soil of the old-field plot.

Of the 2.81×10^6 ug DDT applied to the plot in June, 1976, 2.58×10^6 ug (91.8%) were accounted for as of June, 1977. Screens, ethylene glycol, and run-off contained $40.57 \times 10^{-3}\%$, $0.09 \times 10^{-3}\%$, and $1.18 \times 10^{-3}\%$, respectively, of the DDT applied. Soil was the major sink for DDT in this study, with 91.8% of the applied DDT located there. Most of the remaining 8.2% was probably lost during the formulation and application of the ^{14}C -DDT granules, some was probably lost in organisms (mainly flying insects) leaving the plot, and some was accounted for by experimental error and error in the counting technique.

If residue levels remain constant at the Urbana area for several years, it could be expected that soil and detritus inhabitants and their predators would continue to accumulate DDT residues. This same contamination of a food chain was noted by Dimond et al. (1970) for earthworms (Lumbricus sp. and Allolobophora spp.) and robins (Turdus migratorius) and by Bandy (1972) for the slug (Deroceras sp.) and the short-tailed shrew (Blarina brevicauda).

The possible uptake of DDT by plants in the old-field is another consideration if soil levels of the insecticide

remain constant or increase. Caro (1969) suggested 3 mechanisms by which organochlorine insecticides could be accumulated by plants: 1) translocation through the roots, 2) vaporization of the pesticide from the soil and its subsequent condensation on the aerial portions of the plant, and 3) mechanical transport of contaminated soil particles onto the plant surfaces by wind, rain, or direct contact. He found that the uptake of DDT by the roots of plants is less than that of other organochlorine insecticides, and that it was probably related to the extreme water insolubility of DDT. Ware et al. (1970) found that alfalfa (Medicago sativa) roots and root hairs adsorbed a large amount of ^{14}C -DDT. They concluded that vaporization from contaminated soil to the aerial portion of plants was not a significant route of contamination.

Vaporization of DDT from the soil was reduced to background levels 29 days post application. During these 29 days, vaporization could have played an important role in transporting the DDT from the soil to the aerial portions of the plants. However, after 29 days, the only probably route of transfer would have to have been through the roots. Evidently, even though DDT is accumulated to a lesser extent than other organochlorines, it is accumulated by plants via one method or another. Bandy (1972:74-79) found levels of up to 19.47 ppm ^{36}Cl -DDT in the leaves of grasses, composites, and umbellifers on the

Urbana study area in 1969 as late as 62 days after the original application. The earliest that Bandy found any residues in the plants of the study area was 6 days post application, but in the majority of plant species examined, detectable levels of DDT did not appear in the leaves until at least 23 days post application. Assuming the dissipation of the ^{36}Cl -DDT was equivalent to that of the ^{14}C -DDT and that the majority of the insecticide was absent from the soil surface (and hence, air samples) by 23 days post application, there would have been little available for vaporization and condensation onto the plants. Again, the only probable route of transfer would have been through the roots.

If the soil of an ecosystem is contaminated, eventually all segments of that system will be affected even though some organisms may not be contaminated. As an ecosystem matures, it may become more complex, diverse, and stable (Margalef 1963; Odum 1969). According to Odum's (1969) model, a mature community is characterized by high species diversity, a decreased rate of energy flow per unit of biomass, web-like food chains, and well-defined interspecific relationships such as symbiosis and parasitism. These components of the ecosystem interact and, through feedback mechanisms, promote stability of the system, thus providing protection from external perturbations (Margalef 1963; Odum 1969). Pollutants,

introduced in the ecosystem by either industry or agriculture, exert a profound effect upon the complexity and stability, and thus the maturity, of that system.

Woodwell (1970) reported that pollutants generally, and pesticides specifically, promoted the simplification of ecosystems. Barrett (1968) found a decrease in density and biomass of arthropods important in litter decomposition in a monoculture system treated with the carbamate, Sevin. A decrease in diversity, density, and biomass of primary and secondary consumers also was recorded. Species diversity of the secondary consumers was depressed longer after the application than that of the primary consumers. This difference was attributed to the reduced food supply available to the secondary consumers, lower natality and reinvasion rates, or a combination of the two. The effects of the short-lived insecticide Sevin on community organization were detected long after the toxic residues had disappeared.

With a long-lived insecticide such as DDT, the effects noted by Barrett (1968) may be even greater because the toxic residues are available for a longer period of time. Hence, if soil levels of DDT at the Urbana study area remain constant or increase for at least the 9 years noted by Dimond et al. (1970), the ecosystem may be held consistently in a less complex state and the organization of the old-field community could be

disrupted for a long period of time.

SUMMARY

Carbon-14 DDT was applied in granular form to the surface of a 25-m² plot at a rate of 1 lb/acre. The objectives of this study were to determine the translocation and vaporization of ¹⁴C-DDT into the air and to determine the movement of the pesticide in the surface run-off, ground water, and soil of an old-field ecosystem. Sampling of air, run-off, ground water, and soil was initiated on the first day post application. All samples were analyzed by liquid scintillation spectrometry for ¹⁴C-DDT.

Air samples were collected with a Greenburg-Smith impinger system with ethylene glycol and Chromosorb 102 as the adsorbents and with 16 silk screens, each 841 cm², coated with silicone oil. Carbon-14 DDT residues in the ethylene glycol were highest 3 days post application and decreased rapidly thereafter. Background levels were reached at 29 days post application. No residues were detected in the Chromosorb 102. Residues in silk screens were highest within 2 weeks of application and had declined to background levels by the spring of 1977.

Surface run-off was collected in a sunken 208-l barrel at the lowest corner of the plot. Run-off samples were divided into particulate matter and whole run-off (particulate matter and water). Carbon-14 DDT residues in particulate matter were highest at 6 days post

application, whereas residues in whole run-off were highest at 15 days post application. Within a month, residues in both had declined to background levels.

Ground water was collected with lysimeters at depths of 20, 40, and 60 cm. No ^{14}C -DDT residues were detected in the ground water at any of the depths sampled.

Soil samples were collected from depths of 0-3 cm and 3-6 cm. Carbon- 14 DDT residue levels in the 0-3 cm soil layer were greater than those in the 3-6 cm soil layer by 1 to 2 orders of magnitude which were, in turn, greater than background samples by the same margin. Therefore, ^{14}C -DDT levels in the 0-3 cm layer of the experimental plot were greater than background levels by 2 to 4 orders of magnitude. At the completion of this study in 1977, soil ^{14}C -DDT residues had not declined to background levels.

The presence of ^{14}C -DDT in the air, water, and soil of the old-field plot and the rate of decline of the pesticide was related to time post application, the amount of rain that fell on the plot, and, in the case of the soil samples, ambient temperature. Screens, ethylene glycol, and run-off contained $40.57 \times 10^{-3}\%$, $0.09 \times 10^{-3}\%$, and $1.18 \times 10^{-3}\%$, respectively, of the DDT applied. Soil was the major sink for DDT in this study, with 91.8% of the applied DDT located there. Most of the remaining 8.2% was probably lost during the

formulation and application of the ^{14}C -DDT granules, some was probably lost in organisms (mainly flying insects) leaving the plot, and some was accounted for by experimental error and error in the counting technique.

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Appendix A. ^{14}C -DDT residues in air (ethylene glycol) samples from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Collection Date	DPM	DPM equal to ng DDT/m ³ air ^b
6-20-76 (3) ^a	102.57	3.27
6-25-76 (8)	96.43	3.08
6-28-76 (11)	65.14	2.08
7-01-76 (14)	61.42	1.99
7-04-76 (17)	52.21	1.67
7-07-76 (20)	70.97	2.27
7-10-76 (23)	83.23	2.66
7-13-76 (26)	55.27	1.76
7-16-76 (29)	54.00	1.72
7-19-76 (32)	46.84	1.49
7-22-76 (35)	42.10	1.34
7-25-76 (38)	47.63	1.52
7-28-76 (41)	54.53	1.74
7-31-76 (44)	42.34	1.35
8-03-76 (47)	40.14	1.28
8-06-76 (50)	42.70	1.36
8-09-76 (53)	52.38	1.67
8-12-76 (56)	44.17	1.41
8-15-76 (59)	55.23	1.76
8-18-76 (62)	47.60	1.52
8-21-76 (65)	45.25	1.44

Appendix A (continued).

Collection Date	DPM	DPM equal to ng DDT/m ³ air ^b
8-24-76 (68)	44.70	1.43
8-27-76 (71)	50.28	1.60
8-30-76 (74)	56.73	1.81
9-07-76 (82)	38.92	1.22
9-14-76 (89)	45.30	1.42
9-28-76 (103)	45.70	1.43
10-05-76 (110)	42.00	1.31
10-12-76 (117)	41.74	1.31
10-19-76 (124)	47.05	1.47
10-26-76 (131)	46.43	1.45
11-02-76 (138)	45.92	1.44
11-16-76 (152)	40.00	1.25
11-23-76 (159)	42.88	1.34
4-12-77 (299)	40.86	1.27
4-19-77 (306)	41.06	1.28
5-11-77 (327)	41.00	1.28
5-18-77 (334)	38.31	1.20
6-01-77 (348)	41.51	1.30
6-08-77 (355)	41.32	1.29
6-22-77 (369)	44.68	1.40
6-30-77 (377)	41.32	1.29

^aDays post application.^b \bar{x} background radiation levels were equal to 1.36 ng DDT/m³ air.

Appendix B. Radiation levels in control air (ethylene glycol) samples from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Collection Date	DPM	DPM equal to ng DDT/m ³ air
6-15-76 (-2) ^a	49.17	1.57
9-21-76 (96)	40.36	1.26
11-09-76 (145)	44.82	1.40
4-26-77 (313)	45.35	1.42
5-25-77 (341)	40.94	1.28
6-15-77 (362)	39.70	1.24

^aNumber of days post application.

Appendix C. ^{14}C -DDT residues in air (Chromosorb 102) samples from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Collection Date	DPM	DPM equal to ng DDT/m ³ air ^b
7-13-76 (26) ^a	18.60	0.59
7-16-76 (29)	19.60	0.63
7-19-76 (32)	19.51	0.62
7-22-76 (35)	40.29	1.29
7-25-76 (38)	20.71	0.66
7-31-76 (44)	20.47	0.65
8-03-76 (47)	28.19	0.90
8-06-76 (50)	46.68	1.48
8-12-76 (56)	18.73	0.60
8-15-76 (59)	21.28	0.68
8-18-76 (62)	20.41	0.65
8-21-76 (65)	33.30	1.06
8-24-76 (68)	31.97	1.02
8-27-76 (71)	29.07	0.93
8-30-76 (74)	32.00	1.02
9-07-76 (82)	18.46	0.58
9-14-76 (89)	30.19	0.94
9-28-76 (103)	31.78	0.99
10-05-76 (110)	33.01	1.03
10-12-76 (117)	19.64	0.61
10-19-76 (124)	29.50	0.92

Appendix C (continued).

Collection Date	DPM	DPM equal to ng DDT/m ³ air
10-26-76 (131)	18.54	0.58
11-02-76 (138)	18.40	0.57
11-16-76 (152)	29.00	0.91
11-23-76 (159)	27.89	0.87
4-12-77 (299)	34.21	1.07
4-19-77 (306)	32.90	1.03
4-19-77 (306)	32.00	1.00
5-11-77 (327)	31.51	0.99
5-18-77 (334)	33.95	1.06
6-01-77 (348)	33.62	1.05
6-08-77 (355)	31.24	0.98
6-08-77 (355)	30.52	0.95
6-22-77 (369)	32.70	1.02
6-30-77 (377)	31.77	0.99

^aNumber of days post application.

^bBackground radiation levels were equal to 0.90 ng
DDT/m³ air.

Appendix D. Radiation levels in control air (Chromosorb 102) samples from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Collection Date	DPM	DPM equal to ng DDT/m ³ air
9-21-76 (96) ^a	21.46	0.67
9-21-76 (96)	29.52	0.92
11-09-76 (145)	24.92	0.78
11-09-76 (145)	25.50	0.80
4-26-77 (313)	32.76	1.02
5-25-77 (341)	32.50	1.02
6-15-77 (362)	31.77	0.99
6-15-77 (362)	32.28	1.01

^aNumber of days post application.

Appendix E. Radiation levels in ground water samples from the Urbana study area, Champaign County, Ohio, 1976.

Collection Date	Depth (cm)	DPM ^b
7-16-76 (29) ^a	20	46.97
7-28-76 (41)	20	49.96
	20	48.04
	40	43.49
	60	44.23
7-31-76 (44)	20	42.04
	20	43.96
	20	44.48
	40	226.74
	40	53.44
	40	47.66
	60	49.11
	60	42.25
	60	63.89
8-03-76 (47)	20	43.23
	20	42.17
	20	41.63
	40	41.98
	40	39.35
8-09-76 (53)	20	40.34
	20	37.98
	40	54.03
	40	38.22
	40	37.19
	60	39.83
	60	38.03
8-12-76 (56)	20	38.81
	20	39.95
	20	42.63
	40	41.26
	40	43.22
	60	38.02

Appendix E (continued).

Collection Date	Depth (cm)	DPM
8-16-76 (60)	20	40.25
	20	40.78
	20	105.70
	20	37.69
	40	42.84
	40	41.12
	40	40.51
	60	39.55
	60	38.02
	60	40.12
8-18-76 (62)	20	38.23
	20	38.96
	40	39.48
	60	68.26
	60	41.95

^aNumber of days post application.

^b background radiation levels were equal to 38.64 DPM.

Appendix F. ^{14}C -DDT residues in soil from the Urbana study area, Champaign County, Ohio, 1976.

Collection Date	DPM		DPM equal to PPM DDT ^c		Block Number
	0-3 cm	3-6 cm	0-3 cm	3-6 cm	
6-26-76 (9) ^a		4986.10 ^b		0.16 ^b	8
6-26-76 (9)		8875.20 ^b		0.29 ^b	10
7-02-76 (15)		6279.50 ^b		0.20 ^b	11
7-02-76 (15)		17241.60 ^b		0.56 ^b	12
7-10-76 (23)		47284.80 ^b		1.54 ^b	9
7-10-76 (23)		1921.90 ^b		0.06 ^b	15
7-15-76 (28)		6885.70 ^b		0.22 ^b	4
7-15-76 (28)		5395.10 ^b		0.18 ^b	19
7-23-76 (36)	6270.00	442.50	0.20	0.01	18
7-23-76 (36)	27282.30	621.40	0.89	0.02	1
7-29-76 (42)	93038.50	1689.90	3.02	0.05	5
7-29-76 (42)	4479.70	3347.20	0.15	0.11	6
8-05-76 (49)	912.00	21866.80	0.03	0.71	17
8-05-76 (49)	26731.00	110.90	1.10	0.00	23
8-12-76 (56)	3642.90	1466.50	0.12	0.05	2
8-12-76 (56)	1875.00	2877.50	0.06	0.09	7
8-19-76 (63)	12016.90	2767.40	0.39	0.09	20
8-19-76 (63)	27077.00	167.40	0.88	0.01	24
8-30-76 (74)	22589.00	324.50	0.76	0.01	25
8-30-76 (74)	8561.00	436.50	0.28	0.01	21
9-02-76 (77)	11074.60	658.00	0.36	0.02	14

Appendix F (continued).

Collection Date	DPM		DPM equal to PPM DDT		Block Number
	0-3 cm	3-6 cm	0-3 cm	3-6 cm	
9-02-76 (77)	15240.00	395.60	0.50	0.01	3
9-07-76 (82)	25772.00	1291.00	1.16	0.04	13
9-07-76 (82)	31893.40	849.30	1.53	0.03	16
9-14-76 (89)	13661.20	317.00	0.70	0.01	22
9-14-76 (89)	57231.00	836.00	2.43	0.03	1
9-21-76 (96)	12426.20	1313.00	0.42	0.04	11
9-21-76 (96)	32991.10	8409.00	1.21	0.31	17
9-28-76 (103)	57800.50	243.00	2.61	0.01	8
9-28-76 (103)	85235.20	1359.00	4.67	0.04	13
10-05-76 (110)	15682.40	426.00	0.58	0.01	10
10-05-76 (110)	18061.00	3281.40	1.11	0.11	25
10-12-76 (117)	55550.00	842.00	2.73	0.03	6
10-12-76 (117)	84528.10	1912.40	5.44	0.07	19
10-19-76 (124)	10534.40	291.00	0.56	0.01	21
10-19-76 (124)	16777.40	1561.40	0.77	0.05	15
10-26-76 (131)	9532.40	323.00	0.46	0.01	12
10-26-76 (131)	60198.40	570.10	2.27	0.02	1
11-02-76 (138)	25069.00	409.00	1.32	0.01	24
11-02-76 (138)	30099.50	760.00	1.60	0.02	2
11-09-76 (145)	22399.20	1165.20	1.01	0.04	7
11-09-76 (145)	39667.20	1315.00	1.29	0.04	5
11-16-76 (152)	51567.10	514.00	1.68	0.02	3

Appendix F (continued).

Collection Date	DPM		DPM equal to PPM DDT		Block Number
	0-3 cm	3-6 cm	0-3 cm	3-6 cm	
11-16-76 (152)	22465.00	1059.00	1.02	0.03	14
11-23-76 (159)	29546.00	198.00	1.06	0.01	9
11-23-76 (159)	12575.00	922.10	0.53	0.03	4

^aNumber of days post application.

^bSamples were 0-6 cm.

^c \bar{x} background radiation levels were equal to 35.00 dpm.

Appendix G. ^{14}C -DDT residues in soil from the Urbana study area, Champaign County, Ohio, 1977.

Collection Date	DPM		DPM equal to PPM DDT ^b		Block Number
	0-3 cm	3-6 cm	0-3 cm	3-6 cm	
3-29-77 (285) ^a	9196.10	1404.74	0.48	0.05	20
3-29-77 (385)	38886.89	1119.50	1.84	0.04	16
4-06-77 (293)	36097.63	1397.67	1.79	0.05	22
4-06-77 (293)	22035.93	3889.48	0.98	0.13	23
4-12-77 (299)	25580.42	1095.48	1.27	0.04	18
4-12-77 (299)	11322.46	440.00	0.55	0.01	21
4-19-77 (306)	23977.40	1893.51	1.24	0.06	13
4-19-77 (306)	27817.02	1088.94	1.16	0.04	6
4-26-77 (313)	14409.43	12228.93	0.55	0.40	10
4-26-77 (313)	29281.48	968.17	1.17	0.03	4
5-11-77 (327)	23226.40	663.53	0.75	0.02	11
5-11-77 (327)	33155.37	5459.67	1.08	0.18	5
5-11-77 (327)	23273.50	325.08	0.76	0.01	24
5-11-77 (327)	6750.08	1338.50	0.22	0.04	7
5-18-77 (334)	10675.04	131.00	0.35	0.00	24
5-18-77 (334)	8186.26	211.79	0.27	0.01	14
5-25-77 (341)	23602.50	466.20	0.78	0.02	8
5-25-77 (341)	72172.76	1592.63	2.79	0.05	9
6-01-77 (348)	11423.44	2300.00	0.39	0.07	19
6-01-77 (348)	14312.90	1010.00	0.53	0.03	12
6-08-77 (355)	8796.47	15900.82	0.29	0.52	1

Appendix G (continued).

Collection Date	DPM		DPM equal to PPM DDT		Block Number
	0-3 cm	3-6 cm	0-3 cm	3-6 cm	
6-08-77 (355)	33097.24	1262.69	1.08	0.04	2
6-15-77 (362)	12383.33	3405.47	0.40	0.11	3
6-15-77 (362)	20218.36	876.95	0.66	0.03	15
6-22-77 (369)	39086.18	---	1.31	--	17
6-22-77 (369)	42282.31	1396.00	1.37	0.05	11
6-30-77 (377)	20874.83	438.10	0.68	0.01	6
6-30-77 (377)	30630.73	231.03	1.00	0.01	13

^aNumber of days post application.

^b \bar{x} background radiation levels were equal to 34.76 dpm.

Appendix H. DPM radiation in control soil from the Urbana study area, Champaign County, Ohio, 1976 and 1977.

Collection Date	DPM	
	0-3 cm	3-6 cm
7-23-76	46.83	34.00
7-23-76	42.77	33.26
7-23-76	33.40	32.95
7-23-76	35.32	29.80
7-23-76	32.26	29.35
5-11-77	34.83	33.33
5-11-77	31.71	42.60
6-02-77	35.87	41.26
6-02-77	30.51	54.62
6-30-77	28.43	30.41
6-30-77	24.26	29.26

Appendix I. Rainfall and temperature data from the Urbana
Sewage Treatment Control Plant, Urbana, Ohio,
June 1976.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1	1.17	26	16	21
2	0.91	22	11	17
3		21	12	16
4		24	13	19
5		26	12	19
6		26	10	18
7		27	12	19
8		31	12	21
9		32	13	22
10		32	15	24
11	0.28	31	18	24
12		32	17	24
13		33	19	26
14	T	32	18	25
15		31	18	24
16	0.41	--	13	--
17	1.70	27	14	21
18		30	17	23
19	2.41	26	13	19
20	0.58	26	13	19
21	2.82	23	11	17
22	1.60	23	11	17
23		26	12	19
24	1.30	28	16	22
25	3.68	24	16	20
26	T	26	12	19
27		29	13	21
28		31	17	24
29	1.32	29	18	23
30	2.36	31	16	23
TOTAL RAIN 20.55				
AVERAGE TEMPERATURE		28	14	21

Appendix J. Rainfall and temperature data from the Urbana
Sewage Treatment Control Plant, Urbana, Ohio,
July 1976.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1	0.46	23	12	17
2		26	10	18
3	T	27	15	21
4		28	13	21
5		26	11	19
6		27	13	20
7	0.53	30	15	23
8	0.15	29	16	23
9		28	13	21
10		31	18	24
11		30	21	26
12		33	18	26
13		26	12	19
14		28	14	21
15		35	18	27
16	T	34	21	28
17	1.37	28	10	19
18		25	11	18
19		29	12	20
20		29	15	22
21		29	18	24
22	1.45	29	18	24
23	5.60	29	20	25
24	0.30	33	22	27
25		29	14	22
26		29	15	22
27	0.18	30	17	24
28		31	17	24
29	T	31	18	25
30		29	18	24
31		28	14	22
TOTAL RAIN 10.03				
AVERAGE TEMPERATURE		29	15	22

Appendix K. Rainfall and temperature data from the Urbana
Sewage Treatment Control Plant, Urbana, Ohio,
August 1976.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		28	12	20
2		25	9	17
3		26	9	18
4		25	8	16
5		28	13	21
6	2.01	30	16	23
7	6.05	22	13	18
8	T	20	7	13
9		23	9	16
10		26	10	18
11		29	13	21
12		31	18	24
13	0.10	31	17	24
14	0.08	29	16	22
15	0.71	27	16	22
16	0.10	22	8	15
17		26	8	17
18		25	8	16
19		27	11	19
20		29	12	20
21		28	9	19
22		29	11	20
23		31	13	22
24		31	14	23
25		32	15	23
26	0.89	27	17	22
27	1.78	28	18	23
28		30	19	25
29	0.89	31	11	21
30		23	6	14
31		23	7	15
TOTAL RAIN 12.60				
AVERAGE TEMPERATURE		27	12	20

Appendix L. Rainfall and temperature data from the Urbana Sewage Treatment and Control Plant, Urbana, Ohio, September 1976.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		26	9	18
2	T	23	13	18
3		23	5	14
4	0.89	26	6	16
5		28	8	18
6		22	7	14
7		23	6	14
8		28	6	17
9		32	8	20
10	1.73	28	7	18
11		20	6	13
12		26	8	17
13		30	6	18
14		30	11	21
15		27	4	16
16		28	4	16
17	0.25	22	9	16
18		22	10	16
19		27	8	17
20	0.10	28	8	18
21		28	3	15
22		19	- 1	9
23		21	2	11
24		26	2	14
25		21	2	12
26	0.25	24	7	15
27	1.17	19	13	16
28	0.51	17	8	12
29		18	3	11
30	0.10	20	3	12
TOTAL RAIN 5.00				
AVERAGE TEMPERATURE		24	6	15

Appendix M. Rainfall and temperature data from the Urbana
Sewage Treatment and Control Plant, Urbana, Ohio,
October 1976.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		18	6	12
2		24	4	14
3		25	5	15
4		25	7	16
5		26	7	16
6	0.61	26	13	19
7	0.71	18	4	11
8		11	7	9
9	0.66	15	6	10
10	0.43	11	0	6
11		11	1	6
12		18	2	10
13		23	5	14
14	T	24	2	13
15		18	1	9
16	T	21	0	10
17		14	0	7
18		9	- 7	1
19		14	- 6	4
20	0.64	13	2	7
21	0.89	8	2	5
22		9	- 3	3
23		7	- 3	2
24	1.78	10	- 3	4
25	0.53	13	7	10
26		9	- 1	4
27		8	- 4	2
28	T	4	- 7	- 1
29		7	- 7	0
30		13	- 2	6
31	1.30	8	3	6
TOTAL RAIN 7.54				
AVERAGE TEMPERATURE				
		15	1	8

Appendix N. Rainfall and temperature data from the Urbana
Sewage Treatment and Control Plant, Urbana, Ohio,
November 1976.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1	T	8	- 6	1
2		12	- 4	4
3		13	- 1	7
4	T	11	- 2	5
5		4	- 2	1
6		6	- 2	2
7		11	- 1	5
8	T	6	- 7	0
9	T	1	- 6	- 3
10		12	- 2	5
11	T	6	- 2	2
12		4	- 5	- 1
13		3	- 8	- 3
14		6	- 8	- 1
15		4	- 8	- 2
16		6	- 9	- 2
17		8	- 9	- 1
18		11	- 2	4
19		11	0	6
20		12	- 7	3
21		9	- 6	2
22	0.08	3	- 4	0
23	T	0	- 5	- 3
24	T	0	- 7	- 4
25	T	2	- 5	- 2
26	0.64	13	0	7
27	0.91	14	7	10
28		14	- 3	6
29	0.25	- 2	-14	- 8
30		- 8	-16	-12
TOTAL RAIN 1.88				
AVERAGE TEMPERATURE				
		7	- 5	1

Appendix O. Rainfall and temperature data from the Urbana
Sewage Treatment and Control Plant, Urbana, Ohio,
December 1976.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		- 5	-15	-10
2		1	- 8	- 4
3	T	- 4	-18	-11
4	T	1	-15	- 8
5		- 1	-15	- 8
6		2	-13	- 6
7	0.66	7	- 3	2
8		- 2	-11	- 7
9		- 4	-11	- 8
10		3	- 8	- 3
11	0.03	7	- 4	1
12	T	3	- 2	1
13		4	- 9	- 2
14		- 2	- 9	- 6
15		6	- 7	- 1
16		9	- 4	3
17		1	- 4	- 2
18		7	-11	- 2
19		4	- 9	- 2
20	0.20	12	1	6
21	0.03	2	-13	- 5
22	T	- 6	-14	-10
23	T	- 2	-13	- 8
24		- 2	-16	- 9
25		1	-13	- 6
26	T	2	- 8	- 3
27	0.05	1	-14	- 7
28	T	3	-12	- 5
29	T	- 1	-16	- 9
30	T	- 9	-17	-13
31	0.64	- 4	-24	-14
TOTAL RAIN 1.60				
AVERAGE TEMPERATURE		1	-11	- 5

Appendix P. Rainfall and temperature data from the Urbana Sewage Treatment and Control Plant, Urbana, Ohio, January 1977.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		-12	-24	-18
2	T	-11	-19	-15
3	0.23	- 4	-19	-12
4	0.25	- 2	- 7	- 4
5	0.10	0	- 7	- 3
6	T	- 1	-18	- 9
7	0.18	- 1	-13	- 7
8		- 8	-24	-16
9		- 4	-16	-10
10	0.56	- 8	-15	-11
11	T	-11	-23	-17
12		-13	-22	-18
13		-12	-26	-19
14	0.10	- 2	-24	-13
15	0.51	1	-12	- 6
16	0.03	- 7	-27	-17
17		-22	-31	-26
18	T	-19	-29	-24
19	0.05	- 9	-22	-16
20	T	- 7	-16	-11
21	T	- 3	-15	- 9
22	T	- 4	-17	-11
23		- 7	-18	-12
24	0.10	- 4	-16	-10
25	T	0	- 5	- 3
26	0.08	- 1	- 8	- 4
27	0.05	0	-18	- 9
28	0.05	- 1	-18	- 9
29	T	-12	-24	-18
30		-14	-20	-17
31		-12	-21	-16
TOTAL RAIN 2.29				
AVERAGE TEMPERATURE		- 7	-19	-13

Appendix Q. Rainfall and temperature data from the Urbana Sewage Treatment and Control Plant, Urbana, Ohio, February 1977.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		- 9	-20	-14
2	T	- 7	-21	-14
3	T	1	-17	- 8
4	0.10	3	- 4	- 1
5	0.30	1	-13	- 6
6	0.23	- 6	-23	-14
7		- 6	-23	-15
8		- 7	-27	-17
9		- 3	-27	-15
10		5	- 5	0
11		9	- 2	4
12		11	- 2	4
13	T	6	--	--
14	0.74	6	- 3	2
15	0.23	2	-13	- 6
16	T	- 4	-17	-11
17		- 6	-19	-13
18		- 2	-16	- 9
19	T	3	- 5	- 1
20	0.05	4	- 7	- 1
21	T	- 2	-13	- 8
22		3	-13	- 5
23	0.03	13	3	8
24	0.81	18	4	11
25	0.10	11	3	7
26		10	1	6
27	0.10	9	2	6
28	T	9	- 5	2
TOTAL RAIN 2.69				
AVERAGE TEMPERATURE		3	-10	- 4

Appendix R. Rainfall and temperature data from the Urbana
Sewage Treatment and Control Plant, Urbana, Ohio,
March 1977.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1	T	2	- 4	- 1
2		3	- 9	- 3
3	0.03	6	- 7	0
4	1.70	9	1	5
5	0.56	11	0	5
6		4	- 3	1
7		8	- 4	2
8		5	- 4	0
9		16	0	8
10		18	3	11
11		19	1	10
12	0.13	22	2	12
13	2.16	15	4	10
14	T	14	6	10
15		17	3	10
16		23	3	13
17		12	- 3	4
18	2.24	10	1	6
19	T	12	- 5	4
20	0.51	7	- 2	2
21	T	6	- 3	1
22	0.51	13	1	7
23	0.05	4	- 4	0
24		12	- 5	3
25		6	- 4	1
26		12	- 3	5
27		17	1	9
28	0.86	20	7	14
29	0.33	20	12	16
30		24	13	19
31	T	26	4	15
TOTAL RAIN 9.07				
AVERAGE TEMPERATURE		13	0	6

Appendix S. Rainfall and temperature data from the Urbana Sewage Treatment and Control Plant, Urbana, Ohio, April 1977.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		9	- 3	3
2	2.74	16	0	8
3	6.50	20	2	11
4	0.08	12	4	8
5	0.89	16	2	9
6	0.08	6	- 2	2
7		4	- 3	1
8		18	- 3	8
9		6	- 6	0
10		12	- 1	6
11		27	8	18
12		27	9	18
13		27	11	19
14		27	8	18
15		23	6	14
16		24	6	15
17	0.03	27	9	18
18	0.13	26	9	18
19		31	12	21
20		26	13	20
21	0.15	28	14	21
22	0.56	27	16	21
23	0.76	20	14	17
24	0.30	17	6	11
25	0.25	18	2	10
26	0.41	8	3	6
27	T	16	1	8
28		23	3	13
29	1.78	14	- 2	6
30	T	16	- 2	7
TOTAL RAIN 14.66				
AVERAGE TEMPERATURE		19	5	12

Appendix T. Rainfall and temperature data from the Urbana
Sewage Treatment and Control Plant, Urbana, Ohio,
May 1977.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		21	3	12
2	0.56	26	8	17
3	T	25	10	18
4	3.38	24	13	18
5	0.41	22	14	18
6	T	27	15	21
7	0.81	22	11	17
8		22	2	12
9		21	0	10
10		12	0	6
11		16	2	9
12		22	4	13
13		24	8	16
14		28	12	20
15		29	13	21
16		28	8	18
17		32	13	23
18	0.13	31	17	24
19		32	14	23
20		33	16	24
21		34	13	24
22		32	16	24
23	0.25	29	14	22
24	T	30	13	21
25		32	14	23
26		31	11	21
27		29	9	19
28		33	11	22
29		33	14	24
30		33	11	22
31		32	14	23
TOTAL RAIN 5.54				
AVERAGE TEMPERATURE		27	11	19

Appendix U. Rainfall and temperature data from the Urbana Sewage Treatment and Control Plant, Urbana, Ohio, June 1977.

Date	Rain (cm)	Maximum°C	Minimum°C	Average°C
1		32	16	24
2		29	9	19
3		24	8	16
4		26	8	17
5		31	14	23
6	0.18	27	19	23
7		24	6	15
8	T	19	7	13
9	1.30	21	9	15
10		19	6	13
11	T	24	10	17
12	0.13	24	13	18
13		26	14	20
14	0.05	27	16	21
15		26	13	19
16		29	12	21
17		31	14	23
18		32	17	24
19	0.30	32	17	24
20		29	18	24
21		31	15	23
22		27	14	21
23	T	22	10	16
24		29	13	21
25		31	18	24
26	0.03	31	13	22
27		32	16	24
28	1.22	30	19	24
29	0.05	32	18	25
30		28	13	21
TOTAL RAIN 3.25				
AVERAGE TEMPERATURE		27	13	20