

Compartmentalization of ^{14}C -DDT in an Experimental Old-Field Plot

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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 accumulation of the pesticide in old-field flora and fauna was 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), forest (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, SODERGREN 1973, BIDDLEMAN and OLNEY 1974, GOERLITZ and LAW 1974, HAQUE and FREED 1974) but not for old-field ecosystems. Our objectives were to determine the rate of vaporization and translocation of ^{14}C -DDT into the air, surface run-off, ground water, and soil of an old-field experimental plot. Major plant species included grasses (*Agropyron*; *Dactylis*; *Phleum*; *Poa*); umbells (*Daucus*; *Pastinaca*) and composites (*Achillea*; *Aster*; *Cirsium*; *Solidago*).

METHODS AND MATERIALS

Carbon-14 DDT was applied in granular form to the surface of a 25-m² plot at a rate of 1 lb/acre on 17 June 1976. The plot was located on the Urbana Wildlife Area in Champaign County, Ohio. A trench extending below the water table was dug around the plot on all four sides. The trench was lined with polyethylene sheeting, to prevent horizontal movement of ground water, and backfilled. Sampling of air, run-off, ground water, and soil was initiated on the first day post application. All samples were analyzed for ^{14}C -DDT with a Packard Tri-Carb Scintillation Spectrometer Model 3003. Quench corrections for ^{14}C were determined by channels ratio (HERBERG 1965) and external standardization (HENDEE 1973:211).

Air samples were collected with a Greenburg-Smith impinger system (obtained from the U.S. Public Health Service) with ethylene glycol and Chromosorb 102 (THOMAS and SEIBER 1974) as the adsorbants. Air was collected at ground level by laying

perforated polyethylene hoses over the plot and connecting them to the impinger system. The machine was run 8 h per day for 3 days for a total of 24 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 $2.83 \times 10^{-2} \text{ m}^3$ per minute. Thus, the 24-h samples represented an air volume of 40.75 m^3 . Sixteen silk screens, each 841 cm^2 and coated with $1.6 \pm 1.0 \text{ g}$ silicone oil (Dow Corning 704), (SODERGREN 1974) were also used for the collection of air samples. Each screen was placed on a stainless steel frame and set 20 cm above the ground at a 30° angle above the horizontal. Surface run-off was collected in a sunken 208-l (55-gal), teflon-lined barrel at the lowest corner of the plot. Run-off samples were divided into particulate matter and whole run-off (particulate matter and water). Ground water was collected with lysimeters at depths of 20, 40, and 60 cm. Soil was a Miami silt loam. Cores were collected from depths of 0-3 cm and 3-6 cm. Each core was 2 cm in diameter.

RESULTS AND DISCUSSION

Air Samples - Ethylene Glycol

Carbon-14 DDT residues in the ethylene glycol samples were recorded as ng/m^3 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 background (control) samples. Samples collected through 29 days post application contained significantly ($P < 0.05$) higher ^{14}C -DDT residues than did controls. The residues in the ethylene glycol 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 was similar to the results of other researchers (LICHTENSTEIN and SCHULZ 1959; EDWARDS 1973; PIONKE and CHESTERS 1973).

A rapid loss of the DDT into the atmosphere was obtained possibly because the ^{14}C -DDT was surface applied rather than soil incorporated (CHISHOLM et al. 1950, LICHTENSTEIN and SCHULZ 1961, WHEATLEY 1965, HINDIN et al. 1966). 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.

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. By the time the Chromosorb 102 sampling was initiated, ^{14}C -DDT

residues in the ethylene glycol had declined almost to background levels. Hence, no difference ($P > 0.05$) in ^{14}C -DDT levels was found between background and experimental Chromosorb 102 samples.

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 on screens collected from the study area were significantly ($P < 0.01$) higher than those residues in control (background) screens for both 1976 and 1976 and 1977 combined. No difference ($P > 0.05$) was detected between the 1976 and 1977 residue data from controls nor between the 1976 control and 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 time.

Run-Off Samples

Carbon-14 DDT residues (recorded as parts per billion, ppb) were found to be significantly ($P < 0.01$) higher in particulate matter samples than in whole run-off samples. 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. Residues in whole run-off samples were highest 15 days post application but also declined rapidly and reached background levels by 29 days post application. 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 ^{14}C -DDT granules having been applied to the soil surface or particles of the granules 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 (MC CARTY 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.

Soil Samples

Residues of ^{14}C -DDT (recorded as parts per million, ppm) were significantly ($P < 0.01$) higher in the experimental 0-3 cm soil samples than in the experimental 3-6 cm sections for both 1976 and 1977. Amounts of ^{14}C -DDT present in the soil samples collected in 1976 did not differ significantly ($P > 0.05$) from those collected in 1977. Residues in the experimental soil samples were greater ($P < 0.01$) than those in control (background) samples for both years. Even though amounts of residues did not differ significantly, they declined slightly. A slight upward trend in the amount of DDT (mean ppm per month) in the experimental 0-3 cm samples was observed in 1976. One-way analysis of variance showed no significant differences ($P > 0.05$) among the means of each collection period. Therefore, DDT residues in soil did not differ significantly over the time period sampled.

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 from the data obtained from each segment of this study. (Tables 1 and 2).

TABLE 1.

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 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. The amounts of ^{14}C -DDT in the ethylene glycol samples had declined to background levels by

TABLE 2.
Amounts of ^{14}C -DDT in each "compartment" of the old-field plot at the Urbana study area, Champaign County, Ohio, 1976.

Compartment	dpm ^a			
	Maximum	Minimum	\bar{x}	SD
Air				
Screens	2954.06	26.00	1176.60	1132.93
Ethylene Glycol	102.57	38.92	52.67	15.19
Chromosorb 102	46.68	18.40	26.29	7.66
Run-Off				
Particulate Matter	247.22	33.92	67.00	62.47
Whole Run-Off	408.04	30.73	88.24	120.24
Ground Water	226.74	37.19	48.46	28.78
Soil				
0-3 cm	93038.50	1875.00	26222.46	23124.03
3-6 cm	8409.00	110.90	1228.98	1487.68

^aDisintegrations per minute.

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 m³ of air collected over a period of 216 h, an average collection of 3.8×10^{-3} ug DDT/h. Over the 29-day period, the amount of DDT that could have been collected by the ethylene glycol on the old-field plot had the impinger system been run 24 h a day for the 29 days was determined, by extrapolation, to be at least 2.64 ug DDT. In calculating this estimation, we assumed that the vaporization of DDT per unit time over 24 h was equal to those time periods actually sampled. Based on the assumption that all 16 experimental screens had the capacity to hold the maximum amount of ¹⁴C-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 (13.46×10^3 cm² of 250.00×10^3 cm²). Had screens covered the entire 250.00×10^3 cm² of the plot, the amount of DDT they would have held was determined by extrapolation to be at least 1141.25 ug DDT.

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 in the run-off of the old-field plot.

By the end of this 12-month study, ¹⁴C-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 contained 3.14 cm² of soil. The major portion (94.68%) of the residues detected were in the 0-3 cm soil section. Because the ¹⁴C-DDT had been applied only to that part of the soil, 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 cm² of soil, contained 2.40×10^3 ug DDT, an average of 10.31 ug DDT/cm² of soil. Because there were 250.00×10^3 cm² on the surface of the plot for the application of the ¹⁴C-DDT, by extrapolation, at least 2.58×10^5 ug DDT were present in the soil of the old-field plot.

SUMMARY

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. Silk screens, ethylene glycol air samples, 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.

ACKNOWLEDGEMENTS

This study was supported by the U.S. Department of Energy (contract number AT[11-1]1358), the Department of Zoology at The Ohio State University, the Ohio Cooperative Wildlife Research Unit, and the Ohio Department of Natural Resources, Division of Wildlife. Field and laboratory assistance was provided by J. W. Grimm, J. W. Means, Jr., and P. J. Richie. D. J. Forsyth, Environment Canada, Ottawa, Dr. W. J. Collins and Dr. A. C. Waldron, Department of Entomology, Dr. T. J. Logan, Department of Agronomy, Dr. J. D. Harder, Department of Zoology, The Ohio State University, and Dr. T. A. Bookhout and Dr. G. A. Grau, Ohio Cooperative Wildlife Research Unit, provided advice on various aspects of the project.

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