PESTICIDES IN SOIL

DDT Residues in Forest Floor and Soil After Aerial Spraying, Oregon—1965-68 [1]

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ABSTRACT

One month after aerial application of DDT (12 oz/acre) to an eastern Oregon forest, 3 oz/acre of DDT residues (DDT, its isomers and metabolites—DDD, DDE, p,p′-DDT, and o,p′-DDT) were detected in the forest floor; 3 years later, the DDT content had decreased by more than 50%, and had not leached into the surface mineral soil.

At the time of spraying, water from two streams draining the sprayed area had a total DDT content of about 0.3 ppb. This low concentration decreased rapidly to levels below limits of analytical detection. No effect of the spraying was noted on soil microbial populations, nitrification rate, or amount of nitrate nitrogen in the soil.

Of the 12 oz of DDT applied per acre, about 26% reached the ground surface initially; and over 36 months, about 6% more was brought to the ground in litterfall. Thus, approximately one-third of the sprayed chemical reached the forest floor. The need for more efficient aerial methods of chemical application is evident.

Introduction

Only a few studies have been concerned with residues of DDT, its isomers and metabolites, in the forest environment. Woodwell (*41*) determined DDT residues in a forest in New Brunswick, Canada, which had been sprayed between 1952 and 1958 with a total of 4 lb/acre of DDT. He concluded that the maximum residue persistence time in forest soil would be 10 years and that *o,p′*-DDT was leached into the subsoil. Woodwell and Martin (*42*) reported that DDT residues in heavily sprayed forest soils in Maine and New Brunswick increased over a period of 3 years after final spraying. These authors hypothesized that DDT residues persist in the forest canopy and are carried to the soil by rain and litterfall. Yule (*43*) stated, however, that Woodwell's hypothesis of differential weathering and preferential retention of *o,p′*-DDT by New Brunswick forest soils is untenable. He concluded from a study in the same locality that about 16% of the DDT originally applied still remained in surface soils after almost 20 years, but mainly in the form of the most toxic isomer, *p,p′*-DDT. He further demonstrated that the acidic, highly organic, forest surface soils held these DDT residues unavailable in toxic amounts to soil insects.

DDT residues were also measured in a northern Pennsylvania forest soil, 380 days after aerial spraying at a rate of 0.5 lb/acre (*9*). The only environmental change reported was a significant accumulation of DDT in the forest floor and surface soil. One year after spraying, no measurable increase in DDT residues was noted in fish, crayfish, or stream sediments. Belyea (*4*) studied DDT residues in soils and a related food chain in northern Maine forests. He concluded that DDT would disappear from these soils in 10-12 years. In western Washington, DDT was applied to the surface of a gravelly soil beneath a stand of Douglas-fir [*Pseudotsuga menziesii* (Mirb.) Franco] at 0.5 and 5.0 lb/acre (*29*). Regardless of application rate, less than 1% of the DDT applied leached through the surface soil.

In 1965, an opportunity to further study DDT residues in forests resulted from a spray project conducted by the U. S. Forest Service between June 10 and July 1 to control a serious outbreak of the Douglas-fir tussock moth (*Hemerocampa pseudotsugata* McD.) in eastern Oregon. Helicopters were used to spray 66,000 acres of forest with an insecticide formulation of 0.75 lb of technical grade DDT dissolved in 0.94 qt of hydrocarbon solvent and sufficient No. 2 fuel oil to make 1 gal of solution at 60° F; the application rate was 12 oz/acre.

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A number of public agencies have already conducted surveillance and monitoring activities to determine residues in fish and wildlife, cattle, and forage, and to ascertain public health effects from the spray project (*10,35*). The present study reports observations of the persistence of the applied DDT in the forest floor and soil and some related effects for the first 3 years after the spray project.

The area studied was along a 1-mile transect in the Malheur National Forest of eastern Oregon. Elevation of the area is 5,700 feet. The subhumid continental climate includes dry, warm summers (>100° F maximum temperature) and cold winters (−20° F minimum temperature). The daily range of temperatures in summer is often 40 to 50 degrees and the monthly range may exceed 60 degrees. Annual precipitation averages about 20 inches, one-third to one-half of which is snow. Most moisture available to plants is stored in the soil at the beginning of the growing season, and by midsummer soils become very dry; summer flow in small streams is intermittent.

The soil of the study area, representative of perhaps several million acres of eastern Oregon forest land, is tentatively identified as a member of the Klicker series. This is a well-drained, moderately fine-textured, forested soil developed in residuum from basalt bedrock; small fragments of the basaltic parent material occur throughout the solum, and rock outcrops are common throughout the area. The A horizon, which may contain volcanic ash, ranges in texture from silt loam to silty clay loam, with the percent clay increasing gradually with soil depth. The soils are slightly acidic throughout the solum which ranges in depth from 15 to 35 inches. Permeability is moderate, and surface runoff is medium. Major tree species in the study area which partially determine the amount of litter and duff on the forest floor are ponderosa pine [*Pinus ponderosa* (Laws).], White fir [*Abies concolor* (Gord. & Glend..) Lindl.], and Douglas-fir [*Pseudotsuga menziesii* (Mirb.) Franco].

Sampling Procedures

Five 1/10-acre plots were established along the transect nearest to equidistant points where maximum uniformity of stand composition and density could be found. Within each of the five plots, four sampling points were randomly selected. At each point, the forest floor over a 4-sq-ft area was carefully removed, and 1-qt samples of soil were collected at each of two depths, 0 to 3 inches and 3 to 6 inches, in step-like trenches. Extreme care was taken to avoid contaminating the lower sample with material from above, and all tools were cleaned frequently with acetone. Samples were placed in new 1-qt paper freezer containers, stored during the day in portable cooler chests, and frozen the same day they were collected. Samples of forest floor and soil were collected prior to the spray project, 1 month after the spraying, and at 1-year intervals. Litterfall, throughfall precipitation, and water samples from streams were collected every 6 months.

Eight trays, each with a surface area of 10.9 sq ft, were randomly placed throughout each sample plot to collect litterfall, and four 1-gal containers fitted with funnels having openings approximately 1 sq ft in size were randomly located in each sample plot to collect precipitation. Water samples from the east and west forks of Rattlesnake Creek within 50 feet of their confluence were taken in 1-gal containers, submerged just deep enough to prevent undue disturbance of bottom sediments.

Analytical Procedures

Soil samples were air-dried, ground, passed through a 10-mesh sieve, mixed, and subsampled. For samples intended for microbial analysis, the sieve was cleaned with 30% ethanol and flamed between collections of each sample. Frozen litter and forest floor samples were chopped with dry ice, mixed, and subsampled. All samples, including water, were stored at 0° F until extracted.

EXTRACTION

Soils: A 100-g subsample was extracted with 41:59 hexane:acetone (azeotropic) in a Soxhlet extractor for 16 hours (*27*).

Litter and forest floor: A 25-g subsample was extracted with acetone in a Soxhlet extractor for 16 hours.

Water: Volumes up to 4,000 ml were extracted with hexane in a continuous-cycling liquid-liquid extractor for 16 hours.

CLEANUP

The soil and litter extracts were transferred to separatory funnels. Water was added to form a 2:1 water-acetone solution. The pesticides were partitioned into hexane by shaking with three 100-ml aliquots of hexane (*17*).

The hexane extracts of soil, litter, and water were dried with anhydrous sodium sulfate, evaporated to a small volume (5- to 10-ml), and transferred to a 15-g Florisil column (*40*). The pesticides were eluted from the column with 100 ml of 1:3 dichloromethane:hexane. Dichloromethane was removed by evaporation, and samples were transferred to volumetric flasks for analysis.

ANALYSIS

The concentration of DDE, *o,p′*-DDT, DDD, and *p,p′*-DDT was quantified in a MicroTek 2000 MF gas chromatograph with a 130-mc tritium electron capture detector. This system gave good individual peak resolution at the following retention times: DDE, 5.2 minutes;

o,p'-DDT, 6.8 minutes; DDD, 8.0 minutes; and *p,p'*-DDT, 9.5 minutes. Other operating parameters were:

Column:	Pyrex glass, 180 cm x 2 mm i.d., packed with 5% QF-1 (0.7 of length) and 5% DC-11 (0.3 of length) on 60/80 mesh Gas Chrom Q, preconditioned for 48 hours at 220° C.	
Temperature:	Column	185° C
	Detector	190° C
	Injector	205° C
Carrier gas:	Nitrogen at 30 ml/min	

Minimum residue levels for quantitative determinations were 0.001 ppm for soil, 0.01 ppm for forest floor and litter, and 0.01 ppb for water. Average percent recovery and range for DDT isomers and metabolites were as follows:

FORM OF DDT	SOIL (PPM)	FOREST FLOOR AND LITER (PPM)	WATER (PPB)
DDE	99(92-103)	97(93-100)	99(98-100)
o,p'-DDT	82(71-99)	99(96-100)	98(97-100)
DDD	82(78-91)	85(80-94)	94(93-95)
p,p'-DDT	97(92-100)	94(90-97)	99(98-100)

NOTE: ( ) = range.

CONFIRMATION

It is most necessary to positively identify any apparent DDT determined by gas-liquid chromatography (GLC), especially in samples of materials to which no known DDT application was made. A number of industrial pollutants are similar to DDT in structure and properties and can interfere with the detection or identification of DDT (*19,22,28,30,31*); some naturally occurring plant or soil substances may also be potential sources of analytical error in determining the presence of chlorinated hydrocarbon compounds (*14,20*). To confirm apparent DDT residues in this study, about half the samples were analyzed by GLC with a chloride-specific, microcoulometric detection system (Infotronics Instrument Corp.). This step confirmed that substances with the same retention time as the DDT standards detected by the electron capture detector did contain chlorine, but did not rule out the possible misinterpretation of polychlorinated biphenyls (PCB's) as DDT isomers and metabolites. Therefore, all samples analyzed with the microcoulometric detector were hydrolyzed with alcoholic potassium hydroxide which would chemically alter DDT and DDD, but not PCB's (*17*). Hydrolyzed samples were then re-analyzed by both electron capture and microcoulometric detection systems. DDD, *o,p'*-DDT, and *p,p'*-DDT peaks disappeared after hydrolysis, indicating that PCB's were not present in detectable qualities and that the quantitative measurement of DDT isomers and metabolites by the electron capture detection system was correct.

The mass spectrophotometer provides the most positive means of identifying pesticides in biological samples; but in this study, only a few forest floor samples contained sufficient DDT to use this instrument. Two composite samples of forest floor from two different plots were extracted and purified for this particular analysis. The DDT isomers and metabolites were separated by chromatography of the final hexane extract on 500-μ silica gel H thin layer plates developed with 4:96 benzene:hexane. DDT standards were co-chromatographed on both edges of the 20- by 20-cm plates. After development, a 15-cm strip in the middle of the plate was covered, and the DDT standards were located by spraying the edge of the plate with 0.5% silver nitrate and exposing to UV light for 15 minutes. The *o,p'*-DDT, *p,p'*-DDT, and DDE were scraped from the appropriate section of the center of the plate, extracted from the silica gel with hexane, and analyzed by gas chromatography (electron capture detector). The pesticides separated by the thin layer method had the same retention times as the standards.

Extracts containing the individual pesticides were introduced into a Model CH 7 (Varian Mat. Bmg. H.) mass spectrometer with a direct inlet probe. The mass spectra for *o,p'*-DDT, *p,p'*-DDT, and DDE isolated from the forest floor samples agreed with spectra of appropriate standards and with published spectra (*34*). Sample spectra compared with published PCB spectra (*3*) indicated no PCB's present in the isolated pesticides. All confirmation steps gave positive evidence that the substances isolated and measured were indeed DDT isomers and metabolites and that PCB's were not present in detectable quantities.

MICROBIAL ANALYSIS

The number of bacteria in soil was estimated by plating sterile tap water dilutions of soil on sodium albuminate agar; mold counts were made on plates of peptone-glucose-acid agar (*37*). In each case, five plates were poured of each dilution: 1:50, 1:500, and 1:5,000 for molds; 1:5,000, 1:50,000, and 1:500,000 for bacteria. Mold colony counts were made on plates showing approximately 30 to 100 colonies after 3 days' incubation; major genera were differentiated as mucors, *Penicillium*, and *Aspergillus* after 3 to 7 days. Total bacteria and *Streptomyces* were counted on plates showing colonies in the range of 50 to 300 after 10 to 14 days' incubation; *Streptomyces* were differentiated and their numbers expressed as a percentage of the total count. Incubation was 28° C.

Nitrite and nitrate were determined colorimetrically by the diazotization (*2*) and phenoldisulfonic acid (*18*) methods, respectively. For the nitrification study, 200 ppm N was added as ammonium sulfate to 100 g of soil (oven-dry basis) in 1-pt bottles. Moisture was adjusted to 50% of water-holding capacity, and the bottles were capped with polyethylene film and incubated for 30 days at 28° C. Distilled water sufficient for a 1:5 dilution was then added, and the samples shaken for 15 minutes. After pH was measured, nitrite and nitrate in the supernatant were determined.

Results and Discussion

FOREST FLOOR SAMPLES

A very small amount (0.13 ppm) of "apparent" total DDT residues (*p,p'*-DDE, *p,p*-DDD, *o,p'*- and *p,p'*-DDT) were found in prespray samples of the forest floor (Table 1); this was not confirmed as DDT, its isomers and metabolites by means other than GLC because of its insignificant contribution (<0.01 oz/acre) to the totals found after spraying. One month after spraying, concentration of total DDT in the forest floor was slightly more than 7.5 ppm (Table 1), or 3.08 oz/acre (Fig. 1). Thus, an estimated 26% of the applied DDT (12 oz/acre) reached the forest floor shortly after spraying. DDT residues in the forest floor decreased steadily with time, and at the end of 3 years, more than half the DDT originally added had disappeared. Volatilization, chemical or photochemical degradation, and bacterial decomposition are possible removal mechanisms (*12*).

TABLE 1.—*Concentration of DDT isomers and metabolites in the forest floor before and after aerial spraying, Oregon—1965-68*

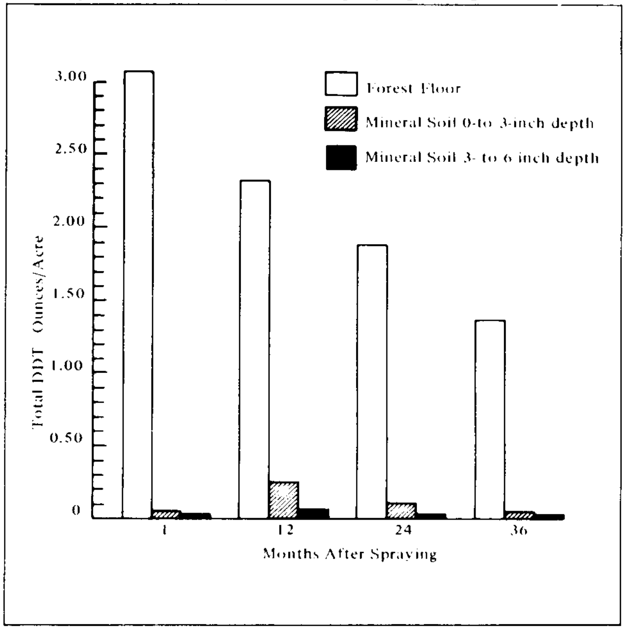
MONTHS AFTER SPRAYING	RESIDUES IN PPM ON A DRY-WEIGHT BASIS [1]				
	p,p'-DDE	*o,p'*-DDT	*p,p'*-DDD	*p,p'*-DDT	TOTAL DDT
0	.008	.025	.008	.089	.130
1	.190	1.294	.348	5.708	7.540
12	.214	.957	.352	3.914	5.437
24	.298	.584	.198	3.332	4.412
36	.096	.473	.114	2.641	3.324

[1] Each value for DDE, DDD, *o,p'*- and *p,p'*-DDT represents the average of duplicate determinations on 20 replicate samples. The relationship between concentrations of all isomers and metabolites and months after spraying is negatively linear and significant at the 5% probability level.

SOIL SAMPLES

DDT did not leach from the forest floor to underlying mineral soil. Apparent total DDT in prespray samples was 0.006 ppm (Table 2, or 0.05 oz/acre (Fig. 1) at the 0- to 3-inch depth; at the 3- to 6-inch depth, total DDT concentration was 0.002 ppm, or 0.02 oz/acre. One month after spraying, these levels had not changed, indicating that the forest floor effectively intercepted the spray solution. One year after spraying, the residue level of total DDT in soil at the 0- to 3-inch depth was 0.26 oz/acre; at the 3- to 6-inch depth, it was 0.05 oz/acre. This small difference in residues 1 month after spraying and 1 year later was attributed to the physical action of soil animals and, most probably, to minor, unavoidable contamination during sampling. DDT has a solubility in water of only about 1 ppb (*7*), and thus, does not leach readily in soil (*16,29,33*). At the end of the second year, total DDT at the upper and lower soil depth has decreased to 0.11 and 0.03 oz/acre, respectively, and by the end of 3 years, was at prespray levels.

FIGURE 1.—*Total DDT in forest floor and mineral soil during 36 months after aerial spraying, Oregon—1965-68*



LITTERFALL SAMPLES

DDT in litterfall totaled 0.73 oz/acre over the 3-year sampling period, about 6% of the original application (Fig. 2). DDT concentration decreased with time at a greater rate than it did in the forest floor and soil (Table 3), suggesting that photochemical decomposition and volatilization may be effective mechanisms of chemical degradation in tree canopies exposed to sunlight. DDT concentration is also reduced in successive litterfall samples because of the constantly decreasing proportion of needles and twigs originally subjected to the spray. The contribution of DDT from litterfall to the forest floor after spraying did not contribute strongly to total amount observed. Total loss of DDT from the forest floor over 3 years amounted to 2.46 oz/acre, more than three times the amount brought down in litterfall over the same period.

THROUGHFALL PRECIPITATION SAMPLES

Additions of DDT to the forest floor by throughfall precipitation were insignificant—0.02 oz/acre for the 3-year period following application (Fig. 2) Concentrations varied with season (summer-fall vs. winter-spring) and, in general, showed a gradual decrease with time (Table 4). DDT concentrations in samples representing the dry summer and fall months were approximately three times greater than those for the wet winter-spring

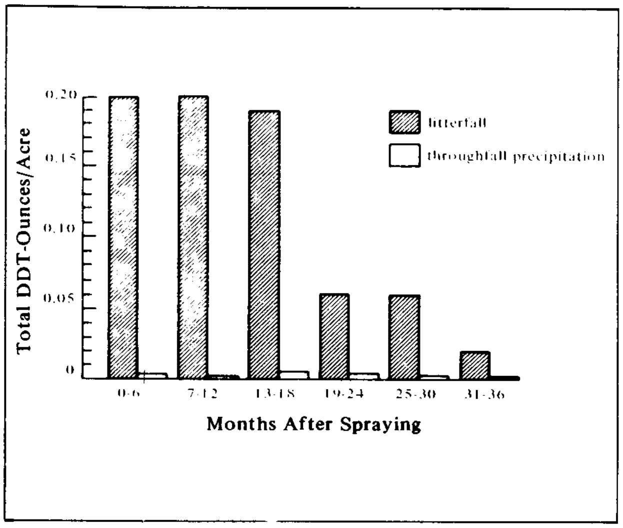
TABLE 2.—*Concentration of DDT isomers and metabolites in surface soil before and after aerial spraying, Oregon—1965-68*

MONTHS AFTER SPRAYING	RESIDUES IN PPM ON A DRY-WEIGHT BASIS [1]				
	p,p'-DDE	*o,p'*-DDT	*p,p'*-DDD	*p,p'*-DDT	TOTAL DDT
	0- to 3-inch depth				
0	.001	.002	—	.003	.006
1	.001	.001	—	.004	.006
12	.003	.005	—	.021	.029
24	.001	.002	—	.009	.012
36	—	.001	—	.005	.006
	3- to 6-inch depth				
0	—	.001	—	.001	.002
1	—	.001	—	.001	.002
12	—	.002	—	.004	.006
24	—	.001	—	.002	.003
36	—	.001	—	.001	.002

NOTE: — = not detected.

[1] Each value for DDE, DDD, *o,p'*- and *p,p'*-DDT represents the average of duplicate determinations on 20 replicate samples.

FIGURE 2.—*Total DDT brought to forest floor in litterfall and throughfall precipitation during 36 months after aerial spraying, Oregon—1965-68*



season. Precipitation samples for the 13- to 18-month period after treatment contained higher DDT concentrations than expected relative to the amount of rainfall for the period and the concentrations found at 6 and 30 months. However, the DDT levels, their seasonal variations, and the total range in concentrations found in this study are consistent with normal climatological variations and similar to those reported for other regions (*1,36,39*).

TABLE 3.—*Concentration of DDT isomers and metabolites added to the forest floor in litterfall after aerial spraying, Oregon—1965-68*

MONTHS AFTER SPRAYING	RESIDUES IN PPM ON A DRY-WEIGHT BASIS [1]				
	p,p'-DDE	*o,p'*-DDT	*p,p'*-DDD	*p,p'*-DDT	TOTAL DDT
0-6	.66	1.57	.44	8.65	11.32
7-12	.19	1.65	.30	8.18	10.32
13-18	.13	1.06	.22	5.71	7.12
19-24	.15	1.07	.26	6.01	7.49
25-30	.09	.62	.13	3.08	3.92
31-36	.07	.47	.10	2.44	3.08

[1] Each value for DDE, DDD, *o,p'*- and *p,p'*-DDT represents the average of duplicate determinations on 20 replicate samples. The relationship between concentrations of DDT isomers and metabolites and months after spraying is negatively linear and significant at the 5% probability level in all cases.

TABLE 4.—*Concentration of DDT added to the forest floor in throughfall precipitation after aerial spraying, Oregon—1965-68*

MONTHS AFTER SPRAYING	PRECIPITATION [1] (MM)	TOTAL DDT RESIDUE [2] (PPB)
0-6	162	.176
7-12	212	.075
13-18	185	.364
19-24	455	.066
25-30	110	.103
31-36	241	.036

[1] Rainfall level extrapolated from monthly climatological data for Burns, Oregon, using Mean Annual Precipitation Isohyetal Map, U. S. Weather Bureau River Forecast Center, Portland, July 1964.

[2] Total DDT residue includes *p,p'*-DDE, *o,p'*-DDT, *p,p'*-DDD and *p,p'*-DDT. DDE residue accounted for an average 6.58% of total DDT residue and showed no statistically significant change with time.

At the end of 3 years, DDT concentrations in throughfall precipitation had decreased appreciably, but still were 5 to 10 times greater than levels found in samples from an untreated forested area in western Oregon; however, the total amount of DDT brought down over this period in throughfall precipitation was negligible compared with that part of the intended application that initially reached the forest floor or was deposited in litterfall. Thus, throughfall precipitation was not a significant factor in determining the fate of applied DDT or in maintaining DDT concentrations in the forest floor.

DDT IN STREAMWATER SAMPLES

Streamwater was monitored in Rattlesnake Creek immediately above the confluence of the east and west forks, both of which flow from the sprayed area. The maximum total DDT concentration found over a period of 3½ years after spraying was 0.277 ppb; this was in a sample taken a few hours after spraying (Table 5). This level was similar to those reported from North Carolina (*15*) and northeastern California (*5*) and less than those from northern Pennsylvania (*9*) and New Brunswick (*44*). Most samples, including those taken 3½ years after the spraying, contained concentrations of DDT near the lower limit of detection (0.01 ppb).

TABLE 5.—*Total DDT content of streamwater flowing from sprayed area—before treatment and during 3 years after treatment, Oregon—1965-68*

DATE	DAYS FROM TIME OF SPRAYING	TOTAL DDT RESIDUES IN PPB	
		RATTLESNAKE CREEK EAST FORK	WEST FORK
1965			
5/24	−30		
6/19	−4	—	—
6/23	1	.104	.277
7/14	21	.031	.022
8/26	64	.028	.015
11/17	147	.014	—
1966			
6/7	349		—
7/19	391	.010	
11/9	505		
1967			
7/4	742	—	—
11/7	869	.032	.010
1968			
7/16	1,131		
11/12	1,251	.010	

NOTE: — = not detected.
Blank = levels of DDT isomers and metabolites less than 0.01 ppb but greater than 0.002 ppb.

Surveillance operations by the Bureau of Sport Fisheries and Wildlife (*25*) indicated that the DDT spraying had little effect on the waters and organisms of Malheur Lake, toward which Rattlesnake Creek flows. Levels of total DDT accumulation in the food chain of Rattlesnake Creek were very low in all components of the sampled community (*8*).

MICROBIAL SOIL PROPERTIES AND NITROGEN RELATIONS

The small amount of DDT in the top 6 inches of mineral soil had no significant effect on microbial populations, soil nitrification rate, or amount of nitrate nitrogen (Table 6). A number of findings similar to these are found in the literature; these reports, however, are based on laboratory studies in which DDT was added to soil at extremely high rates—50-2,000 lb/acre (*13,21,24,26,32*). Effects of pesticide residues on microbes in agricultural soils are usually negligible when the chemicals are used at recommended field rates (*6, 11,23*). Such field rates were usually much greater than those encountered in this study. Thus, it may be concluded that the small amount of total DDT residue in soil found after low-volume aerial spraying to control insects is not hazardous to soil microbes or their role in maintaining soil fertility.

FATE OF AERIALLY APPLIED DDT

Of a total aerial application of 12 oz of DDT per acre, 26% reached the forest floor initially, 6% was brought to the forest floor in litterfall over a 3-year period, and a fraction of 1% of the total was washed from the tree canopy over 3 years (Table 7). Thus, about one-third of the total application reached the forest floor.

TABLE 6.—*Soil microbial populations, nitrate nitrogen content, and nitrification rate before treatment and during 24 months after aerially spraying with DDT at 12 oz/acre, Oregon—1965-68*

VARIABLE AND UNIT OF MEASUREMENT	SOIL DEPTH (INCHES)	MONTHS AFTER AERIAL SPRAYING [1]			
		0	1	12	24
Total bacteria—millions/g of soil	0-3	2.51	2.81	3.46	7.58
	3-6	1.83	2.05	4.04	4.51
Streptomyces—percent of total bacteria	0-3	15.25	23.63	27.25	63.81
	3-6	13.13	19.50	35.56	34.00
Total molds—thousands/g of soil	0-3	338.19	302.88	464.69	168.31
	3-6	93.88	33.88	323.31	35.06
Penicillia—percent of total molds	0-3	78.23	94.46	95.06	62.72
	3-6	77.10	68.45	82.78	68.45
Nitrate nitrogen in soil—ppm	0-3	58.00	40.94	7.00	17.50
	3-6	5.81	4.31	1.63	2.13
Nitrification of added ammonium sulfate—percent of total added	0-3	9.59	−.56	−.50	−.17
	3-6	.27	.04	−.26	−.13

[1] Results of regression analysis indicated no significant relation between soil DDT content and any of the variables measured at the 5% probability level.

TABLE 7.—*Total DDT deposited on the ground surface (forest floor) during 3 years after aerially spraying of DDT at 12 oz/acre, Oregon—1965-68*

SOURCE OF DDT AT GROUND SURFACE	TOTAL DDT	
	OZ/ACRE	PERCENT OF TOTAL APPLIED
Initial deposit from spraying	3.08	25.66
Total deposit during 3 years from:		
Litterfall	.74	6.17
Throughfall precipitation	.02	.01
Total, all sources	3.84	31.84

Because this study did not include direct measurement of the amounts of DDT that reached the forest canopy, the extent of chemical loss from drift during spraying or by volatilization or degradation in the canopy after spraying cannot be assessed. In a study in Arizona, less than 50% of aerially sprayed insecticides were deposited on the agricultural target during summer months (*38*); in the same study, the distance from the spray aircraft to the target was inversely correlated with amount of on-target chemical application. Aircraft spraying forest lands must fly at far greater heights than those operating over level agricultural fields. Thus, the comparatively low amount of on-target application suggested by the present study is not surprising. This finding reaffirms that efficient methods of aerial spraying must be developed in order to avoid great loss of chemical to nontarget areas.

See Appendix for chemical names of compounds discussed in this paper.

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An alternate example could be the requirement for a plant to convert a non-toxic chemical into a toxic one. Plants which are sensitive to 2,4-DB have the ability to rapidly degrade the non-active 2,4-DB to active 2,4-D. Legume plants are much less efficient at making this conversion and therefore are spared from the injury which would be caused by the conversion. Douglas fir is tolerant to 2,4,5-TB because it cannot convert the compound to 2,4,5-T rapidly enough to cause injury. Pine trees can make the conversion and are injured.

We should recognize, however, that all of the above mechanisms of selectivity are not absolute. Each of these can be affected by genetic and environmental factors. The selectivity of dinoseb in peas, mentioned above, can be destroyed by mechanical damage from equipment passing over the peas or from wind storms which may cause blowing sand to damage the waxy cuticle on the leaves. Absorption into the plant may be influenced considerably by the addition of wetting agents in the solution. Temperature can have a very marked effect on the movement and the activity of the herbicide within a plant. This may result in reduced selectivity between two plant species.

From the mouthwash user to the grower applying atrazine to his corn, the selective action of chemicals can be highly beneficial to society. More knowledge must continually be sought in order to most effectively use the selective properties of our modern herbicides.

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PERSISTENCE OF SOME CHEMICALS IN PACIFIC NORTHWEST FORESTS

Introduction

"Persistence" is the tendency of a chemical compound to remain in an unaltered form. Results of research on persistence of DDT, phorate, and a number of herbicides indicate that environmental factors, characteristics of application method and rate, and nature of the chemical used are all important factors in determining length of persistence. Persistence of a given chemical cannot be assumed for one area on the basis of results from use in dissimilar localities.

In a paper included in the proceedings of this course, Norris (7) defined "persistence" as the tendency of a chemical compound to remain in an unaltered form. He also covered in some detail the various factors which determine persistence.

In this presentation I will summarize observations of persistence of several insecticides and herbicides used operationally in Pacific Northwest forests. Results of these field and laboratory studies are most valuable in assessing the environmental effects of economic chemicals used to increase forest production.

Persistence of Two Insecticides in Oregon Forests

DDT

The largest all-helicopter insect spray project ever conducted by the U.S. Forest Service was carried out between June 10 and July 1, 1965, in eastern Oregon. To control a serious outbreak of the Douglas-fir tussock moth (*Hemerocampa pseudotsugata* McD.), 66,000 acres of forest were sprayed with DDT at the rate of 12 ounces per acre.

Since shortly before the spraying, we have studied persistence of the aerially applied DDT in the forest floor and soil (11). A very small amount (0.13 ppm) of "apparent" DDT was found in prespray samples of the forest floor (Table 1). One month after spraying concentration of DDT in the forest floor was slightly more than 7.5 ppm. Based on weight of the

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Table 1. – Concentration of total DDT in the forest floor before and after aerial spraying.

Months After Spraying	Total DDT (ppm)
0	0.130
1	7.540
12	5.437
24	4.412
36	3.324

forest floor, this concentration amounted to 3.08 ounces per acre. Thus, about 26 percent of the intended DDT application of 12 ounces per acre reached the forest floor shortly after spraying.

DDT in the forest floor decreased steadily with time. At the end of the third year after spraying, more than half the DDT originally added to the forest floor had disappeared. Mechanisms of removal of DDT from the forest floor may include volatilization, chemical or photochemical degradation, or bacterial decomposition.

DDT did not leach from the forest floor to underlying soil (Table 2). Concentration of apparent DDT in prespray samples was 0.006 ppm at the 0-3-inch depth and about 0.002 ppm at 3-6 inches. One month after spraying, these DDT levels had not changed, indicating that the forest floor effectively intercepted the spray solution. One year after spraying, DDT in the surface soil was at 0.029 ppm and in the lower soil, 0.006 ppm. This small increase is attributed to the physical action of soil animals and, most probably, to minor, unavoidable contamination during sampling. DDT has a solubility in water of only about 1 ppm (2, 8, 9).

Table 2. – Concentration of total DDT in surface soil before and after aerial spraying.

Months After Spraying	Total DDT (ppm)
0-3-inch depth	
0	0.006
1	.006
12	.029
24	.012
36	.006
3-6-inch depth	
0	.002
1	.002
12	.006
24	.003
36	.002

At the end of the second year, DDT had decreased to 0.012 and 0.003 ppm, respectively, in the upper and lower soil depths. By the end of 3 years, DDT in mineral soil was at prespray levels.

DDT concentration in litterfall decreased with time at a greater rate than it did in the forest floor and soil (Table 3). Photochemical decomposition and volatilization may be effective mechanisms of chemical degradation in tree canopies exposed to sunlight. DDT concentration is also reduced in successive litterfall samples because of the constantly decreasing proportion of needles and twigs originally subjected to the spray.

Table 3. – Concentration of total DDT added to the forest floor in litterfall after aerial spraying.

Months After Spraying	Total DDT (ppm)
0-6	11.32
7-12	10.32
13-18	7.12
19-24	7.49
25-30	3.92
31-36	3.08

The input of DDT to the forest floor from litterfall after spraying did not contribute strongly to total amount observed. Total loss of DDT from the forest floor over 3 years amounted to 2.46 ounces per acre, more than three times the amount brought down in litterfall over the same period.

Additions of DDT to the forest floor by throughfall precipitation were insignificant--0.02 ounce per acre for the 3-year period following application. Concentrations varied with season (summer-fall vs. winter-spring) and, in general, showed a gradual decrease with time (Table 4). DDT concentrations in samples representing the dry summer and fall months were approximately three times greater than those for the wet winter-spring season. Precipitation samples for the 12- to 18-month period after treatment contained higher DDT concentrations than expected relative to the amount of rainfall for the period and the concentrations found at 6 and 30 months. However, the DDT levels, their seasonal variations, and the total range in concentrations found in this study are consistent with normal climatological variations and similar to those reported for other regions (1, 10, 13).

Table 4. -- Concentration of total DDT added to the forest floor in throughfall precipitation after aerial spraying.

Months After Spraying	Total DDT Residue (ppb)
0-6	0.176
7-12	.075
13-18	.364
19-24	.066
25-30	.103
31-36	.036

At the end of 3 years, DDT concentrations in throughfall precipitation had decreased appreciably but still were five to 10 times greater than levels found in samples from an untreated forested area in western Oregon (unpublished data). But the total amount of DDT brought down over this period in throughfall precipitation is infinitesimal compared with that part of the intended application that initially reached the forest floor or was deposited in litterfall. Thus, throughfall precipitation was not a significant factor in determining the fate of applied DDT or in maintaining DDT concentrations in the forest floor.

Of a total aerial application of 12 ounces of DDT per acre, 26 percent reached the forest floor initially, 6 percent was brought to the forest floor in litterfall over a 3-year period, and a fraction of 1 percent of the total was washed from the tree canopy over 3 years (Table 5). Thus, about one-third of the total application reached the forest floor.

In Arizona, less than 50 percent of insecticides aerially applied during the summer months was deposited on-target in agricultural spraying. The distance from the spray aircraft to the target was shown to be inversely correlated with amount of on-target chemical application (12). Aircraft spraying forest lands must fly at far greater heights than those operating over level agricultural fields. Thus, the comparatively high amount of DDT reaching the forest floor is not surprising. This finding reaffirms that efficient methods of aerial spraying must be developed if we are to avoid undue loss of chemical to nontarget areas.

Table 5. -- Total DDT deposited on the ground surface (forest floor) over 3 years after aerially spraying DDT at 12 ounces per acre.

Source of DDT at Ground Surface	Total DDT	
	Ounces per acre	Percent of Total Applied
Initial deposit	3.08	25.66
Total deposit over 3 years from:		
Litterfall	.74	6.17
Throughfall precipitation	.02	.01
Total, all sources	3.84	31.84

PHORATE

Phorate (Thimet) is a highly toxic systemic organophosphate insecticide used heavily in cotton production and also on some truck crops in southwestern United States. This chemical controls insects on cotton for up to 5 months, but residues in soil have disappeared long before harvest. Climatic conditions in southwestern United States, of course, are greatly different from those in the Douglas fir region of the Pacific Northwest.

Phorate was applied broadcast on the forest floor at rates of 1, 10, and 100 pounds per acre in a stand of young Pacific silver fir at each of two western Oregon locations in May 1966 (3). Persistence of phorate and its metabolites (measured as phosphorothiolate sulfone) was determined at the end of each of the first two growing seasons after application. The purpose of this experiment was to determine whether phorate would protect young trees against aphids and whether residue persistence was similar to that in warmer climates.

Mean concentrations in the forest floor after 6 months were 2.04-, 0.58-, and 631-ppm phorate and 2.78-, 26.56-, and 238-ppm metabolite for the 1-, 10-, and 100-pound-per-acre rates, respectively. Complete degradation was quite rapid at the two lower rates as indicated by total residues of 8.40 and 9.98 percent of chemical applied for the forest floor and surface 12 inches of soil combined. At the 100-pound rate, however, 98 percent of the total chemical applied was still present in the forest floor and soil as phorate or its metabolites. Some downward movement had taken place, but less than 4 percent of the total residue was found below the 3-inch soil depth. After 18 months, measurable levels of both phorate and its metabolites were still present in the forest floor and soil. At the highest rate of application, the mean total residue was 4.50 pounds per acre.

This study shows again that we cannot predict the behavior and persistence of an economic chemical in a forest situation from previous experience under agricultural conditions. Residues in this forest study persisted beyond the first growing season probably due to a combination of factors. During the summer months, microbial activity is low because of dry conditions in the forest soil. Chemical degradation did take place at lower rates, but the highest rate of chemical applied was sufficiently toxic essentially to stop the already low level of microbial metabolic activity. By the time the soil moisture supply increased with fall rains, soil temperatures had already dropped to less than optimum and microbial activity remained low until the following spring.

Persistence of Herbicides

Four herbicides are commonly used in the Pacific Northwest to control unwanted vegetation: 2,4-D, amitrole, 2,4,5-T, and picloram. These chemicals are listed in order of persistence. All of them are degraded in the forest floor but at different rates (6).

In red alder forest litter, 2,4-D degrades most rapidly of all (Figure 1). In laboratory studies, after 35 days' incubation, only 6 percent remained of a total application of 2,4-D at the rate of 2 pounds per acre and only 5 percent remained of a 4-pound-per-acre application. The rate of 2,4-D degradation is slowed somewhat when either 2,4,5-T or picloram is applied with 2,4-D, but after 35 days, total degradation is the same as for 2,4-D alone.

Amitrole also degrades rapidly. After 35 days' incubation, only 20 percent of a 2-pound-per-acre application of amitrole remained in forest floor material. Persistence of amitrole, was not affected when 2,4-D was applied concurrently.

Degradation of 2,4,5-T is somewhat slower than that of 2,4-D or amitrole, but after 120 days, only 13 percent of a 2-pound-per-acre application and 18 percent of a 4-pound-per-acre application remained in red alder forest floor material.

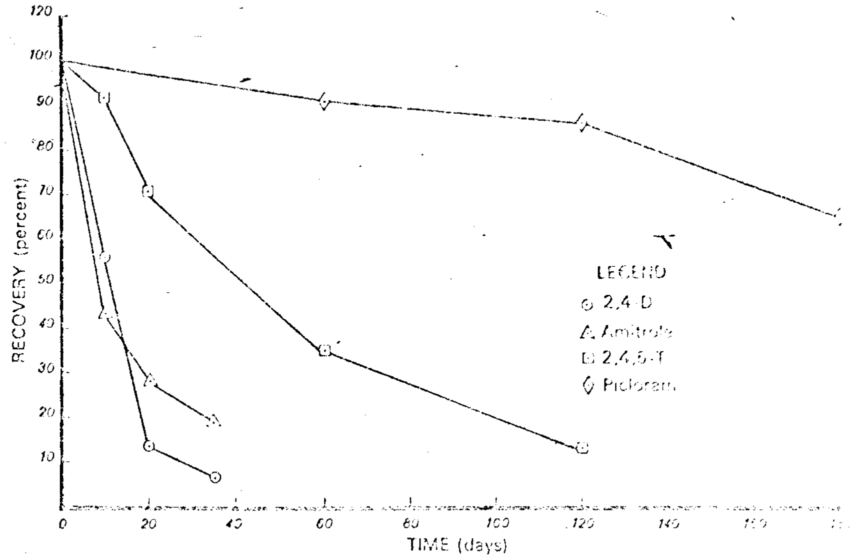

Figure 1. -- Recovery of 2,4-D, 2,4,5-T, amitrole, and picloram from red alder forest floor material. Source: Norris 1970(a).

Picloram (trademark Tordon) is a relatively new herbicide of considerable potential use in controlling unwanted vegetation. It is normally applied at a lower rate than 2,4-D or 2,4,5-T but frequently in combination with them. When picloram was applied at 0.5 pound per acre with and without 2,4-D at 2 pounds per acre, there was no significant difference in persistence of picloram (6). After 180 days, however, more than 60 percent of the initial application of picloram remained in the forest floor.

Results of these laboratory studies have been confirmed by field studies of operational spraying. A number of monitoring observations in western Oregon and Washington have indicated that when forest lands are sprayed with herbicides during the spring growing season measurable quantities of herbicides are not present in streamwater during the first fall rains. Such rains did not introduce measurable amounts of herbicides in the streams flowing through the small scattered treatment units resulting from operational vegetational control measures. These findings are true for 2,4-D, 2,4,5-T, and amitrole. Even in cases where a fairly large proportion of a single watershed is treated with herbicides and where the treatment unit is oriênted for a considerable distance along the stream, no measurable amount of herbicide was found after the first fall rains (4). Thus, unless a heavy application of chemical is made directly into the stream, the major potential for stream contamination from herbicide use in the forest is from heavy rain or soil movement resulting in overland flow of water and sediment shortly after application. Such conditions occur only rarely.

Picloram is sometimes applied with phenoxy herbicides in the summer for brush control on powerline rights-of-way over forest lands. The effect of such summertime spraying on streamwater quality after the first few fall storms has been studied.

Runoff of picloram and phenoxy herbicides can occur after the first heavy autumn rains if the chemicals are applied in mid- or late summer. The greatest potential for herbicide runoff appears when early fall storms are sufficiently intense to cause overland flow rather than infiltration of water. The resulting amount of stream contamination is determined largely by the proportion of the watershed that is treated (5). In our studies, contamination was very low—a maximum of 6 ppb was found in only one sample.

Summary

In an eastern Oregon forest, more than half of the DDT originally reaching the forest floor after aerial spraying had disappeared at the end of the first 3 years after spraying. None of the applied DDT was present in the mineral soil at the end of 3 years.

Under western Oregon conditions, residues of the highly toxic systemic organophosphate insecticide phorate degraded quite rapidly when application rate was either 1 or 10 pounds per acre. However, at an application rate of 100 pounds per acre, about 93 percent of the chemical was still present in the forest floor and soil after 6 months. At the 100-pound-per-acre rate, a total residue of 4.50 pounds per acre was still present after 18 months. Findings from this study under forest conditions are entirely at variance with those for the same chemical used on agricultural crops in a warmer climate.

Of four herbicides studied, 2,4-D degrades most rapidly. Amitrole also degrades rapidly, but 2,4,5-T and picloram are of somewhat longer persistence. Studies of streamwater following first rains after application of these chemicals indicate, however, that 2,4-D, 2,4,5-T, and amitrole residues are not available for transport to streamwater after having weathered over one summer.

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