

# SOILS

DDT Residues in Forest Floors and Soils of Western Oregon, September-November 1966

Duane G. Moore and Bobby R. Loper 1

#### **ABSTRACT**

Between 1945 and 1965, 1.82 million hectares, or about 17 percent of the total commercial forestland in Oregon, were treated with 2.02 million kg DDT. Detectable residues of this insecticide might be present in forest soils, even those which have never received a direct application of insecticide. Forest floor and mineral soil samples were collected along four east-west transects across the Coast and Cascade Ranges. DDT residues were found in all samples, even though all but one site had never received a direct application of insecticide. In the Coast Ranges, mean concentrations of  $\Sigma DDT$  in forest floor samples were 0.049 ppm at the coast and 0.047, 0.064, 0.075, and 0.119 ppm at 16, 32, 48, and 64 km inland, respectively. Mean residue levels in the surface layers of mineral soil were much lower, 0.009 ppm and 0.006 ppm in the 0 to 7.5-cm and 7.5 to 15-cm depths, respectively.

Sampling sites along the Cascade Range transects were selected on the basis of elevation except that the eastern site of each transect was located 16 km east of the crest of the Cascades. Residue concentrations in forest floor samples were three to four times higher than in the Coast Ranges, but were still below 0.50 ppm. In general, EDDT levels increased with increasing elevation up to 1,372 meters and then decreased quite sharply east of the crest. Variations can be explained on the basis of total rainfall distribution and by transect location relative to agricultural and metropolitan centers.

## Introduction

Intensified forest management is essential to meeting the ever-increasing demands for forage, recreation, timber, water, and of wildlife on our forested lands. The inten-

sified use of our forest resources necessitated increased use of pesticides in insect, disease, and brush control programs. As a direct result, synthetic organic pesticides have become a chemical fact of life in forest protection and cultural practices.

Many pesticides in current use are rapidly detoxified in the forest environment. DDT, however, is highly resistant to degradation and may persist for extended periods of time. DDT is not currently registered by the U.S. Environmental Protection Agency (EPA) for use in forestry, but it has an extensive history of use and may be proposed to control specific pest outbreaks in the future. The present study was conducted to determine the levels and distribution of DDT residues in the forest floors and soils of western Oregon.

DDT was first applied to Oregon forests in 1945 to control an outbreak of the western hemlock looper in a 931-hectare (ha) aerial spray project near Cannon Beach (Fig. 1). Between 1945 and 1965, 1.82 million hectares, about 17 percent of the total commercial forestland in Oregon, were treated with 2.02 million kg DDT in various spray projects (Table 1). Applications were heaviest between 1949 and 1958 when 1.77 million hectares were treated in various spruce budworm spray projects.

Since 1958, about 104,682 ha have been treated with DDT, the latest project being carried out on 64,822 ha during 1974 under a Section 18 specific exemption from registration requirement granted to the Forest Service, U.S. Department of Agriculture, by the EPA.

DDT enters the forest floor and soil from direct application, drift of spray materials, rain washing through treated canopy, and litterfall, or in precipitation or dust deposited in unsprayed areas. Information on the extent of actual and potential environmental contami-

<sup>&</sup>lt;sup>1</sup> Forest Service, U.S. Department of Agriculture, Forestry Sciences Laboratory, 3200 Jefferson Way, Corvallis, Oreg. 97331. The use of trade, firm, or corporation names in this publication is for the information and convenience of the reader. Such use does not constitute an official endorsement or approval by the U.S. Department of Agriculture of any product or service to the exclusion of others that may be suitable.

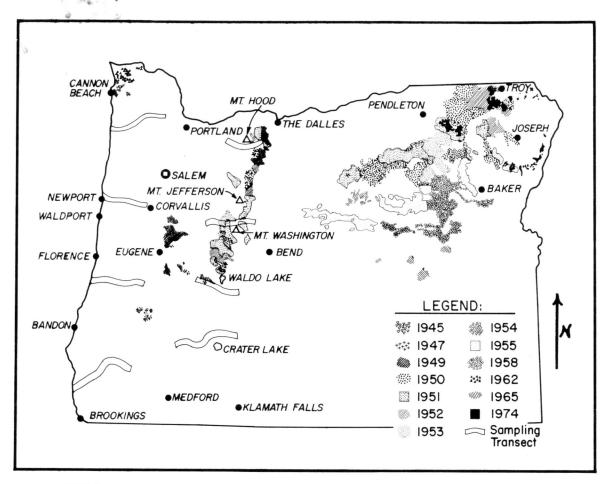


FIGURE 1. Forest insect aerial spray projects in Oregon, and location of sampling transects

TABLE 1. Application of DDT to forests in Oregon, 1945-74

		DDT A	APPLIED, KG
YEAR	HECTARES	BY YEAR	CUMULATIVE
1945	931	1,043	1,043
1947	5,666	6,350	7,393
1948	1,700	1,905	9,298
1949	109,508	122,742	132,040
1950	367,213	411,590	543,630
1951	319,986	358,656	902,286
1952	211,206	236,730	1,139,016
1953	149,815	167,920	1,306,936
1954	27,397	30,708	1,337,644
1955	251,270	281,636	1,619,280
1958	331,034	371,038	1,990,318
1962	13,152	7,371	1,997,689
1965	26,710	22,453	2,020,142
1974	64,822	54,492	2,074,634
TOTALS	1,880,410	2,074,634	

nation with pesticide residues in the forest is limited. Only a few studies have been concerned with  $\Sigma$ DDT. From a study conducted in a forest in New Brunswick, Canada, Woodwell (26) concluded that DDT residues would persist for a maximum of 10 years and that o,p'-DDT was leached into the subsoil. The forest had been sprayed with a total of 4.48 kg/ha of DDT between 1952 and 1958. Woodwell and Martin (27)

reported that DDT residues in soils of heavily sprayed forests in Maine and New Brunswick had increased over a period of three years after the last application. They hypothesized that DDT residues persisted in the forest canopy and were carried to the soil by rain and litterfall.

A study in the same locality refuted Woodwell's suggestion that DDT residues were subject to differential weathering and preferential retention of o,p'-DDT (28). Yule reported that about 16 percent of the DDT originally applied still remained in surface soils after almost 20 years, and that the main component of the residue was p,p'-DDT. He also demonstrated that these DDT residues were unavailable to soil insects in toxic amounts (28).

The only environmental change reported from a study in northern Pennsylvania was a significant accumulation of DDT in the forest floor and surface soil (6). One year after aerial spraying at a rate of 0.56 kg/ha, no measurable increase in DDT residues was noted in fish, crayfish, or stream sediments. Belyea (5) measured DDT residues in soil and a related food chain in

northern Maine forests and concluded that the residues would disappear in 10–12 years. Riekerk and Gessel (18) reported that regardless of application rate, less than 1 percent of the DDT applied to the surface of a gravelly soil beneath a stand of Douglas fir (Pseudotsuga menziesii (Mirb.) Franco) leached through the surface soil over an 18-month period during which 100 cm of rainfall was recorded.

Tarrant et al. (23) conducted a long-term study of the behavior and fate of DDT in a forest environment in eastern Oregon after a 1965 aerial spray project. DDT residues were measured in throughfall precipitation, fresh litterfall, the forest floor and mineral soil, and in water samples from two streams draining the sprayed area. They reported that DDT residues in the forest floor had decreased by more than 50 percent after three years and had not leached into the mineral soil. Movement of DDT from the forest canopy to the forest floor was small and decreased with time. **SDDT** content of stream samples was only 0.3 ppb at the time of spraying, and this low concentration decreased rapidly to levels below the limit of detection. Only about one third of the applied chemical, however, was accounted for; 26 percent reached the forest floor initially, and another 6 percent reached the forest floor in litterfall over a three-year period. Their study did not measure the amount of DDT that had reached the forest canopy, and the extent of chemical loss from drift during spraying could not be assessed. It was assumed that a significant amount of chemical was lost to the atmosphere as drift during application or by volatilization from the canopy after spraying.

Other studies have reported the loss of significant amounts of insecticide to the atmosphere (8, 11). Acree et al. (2) demonstrated that there is significant codistillation of DDT with water at 25° to 35°C. Middleton (15) reported DDT residues as high as 1.71 g/1,000 cu. meters in the air adjacent to application sites. Two studies on the presence of organochlorine pesticides in rainwater were conducted in England, one in an agricultural area (24) and the other in London (1). Both studies reported detectable levels of p,p'-DDT and p,p'-DDE. Antommaria et al. (3) reported minimum concentrations of up to 1.14 µg/1,000 cu. meters for p,p'-DDT associated with suspended particulate matter in Pittsburgh, Pennsylvania, while Tabor (22) reported higher minimum concentrations of airborne DDT, up to 23  $\mu$ g/1,000 cu. meters, mean of 4  $\mu$ g/1,000 cu. meters, in agricultural communities.

Substantial use of DDT in the past, and research on deposition, persistence, and leaching characteristics of DDT, indicate that significant amounts of the insecticide may be present in the atmosphere. Rainfall and deposition of dust could result in detectable residue

levels in forest soils, even those which have never received a direct application of DDT.

# Sampling, Locations and Methods

The general areas sampled were the heavily forested sections of western Oregon. Sampling transects were selected in two subregions, the Coast and the Cascade Ranges. Four east-west transects were sampled in each subregion to cover western Oregon from north to south. In the Coast Ranges, sampling sites were located 0, 16, 32, 48, and 64 km inland along each east-west transect. Elevation was used to locate sampling sites in the Cascade Range. Sampling sites were located at 457, 762, 1,067, and 1,372 meters above sea level along each east-west transect across the Cascades. Sampling sites at the highest elevation, 1,372 meters, were approximately at the crest of the Cascades, and a fifth site on each transect was located 16 km east of the crest. This sampling site is in the rain shadow of the Cascade Range on each transect. Precipitation tends to decrease approximately 25 mm each 1.6 km eastward of the crest.

At each sampling site along the eight transects, four sublocations were sampled north, east, south, and west at 10, 20, 15, and 5 meters, respectively, of an arbitrarily located central point. At each point, the forest floor over a 0.4-sq. meter area was carefully removed, and approximately 1-kg samples of mineral soil were collected at each of two depths, 0 to 7.5 cm and 7.5 to 15 cm. Soil samples were collected by digging shallow soil pits and sampling horizontally into a freshly cleaned vertical face at the desired depth using a square-tipped, flat spade.

Extreme care was taken to avoid contaminating the lower sample with material from above, and all tools were cleaned between samples with acetone. Forest floor samples were placed in new, 10-kg heavyweight Kraft bags, and soil samples were placed in standard soil sample bags. All samples were stored during the day in portable cooler chests and frozen the same day they were collected. In all, 160 sublocations were sampled along the eight transects, resulting in a total of 160 forest floor samples and 320 soil samples. Sampling was accomplished over a three-month period, September–November 1966.

## Analytical Procedures

Soil samples were air-dried, ground to pass a 10-mesh sieve, mixed, and subsampled. Forest floor samples were essentially dry when collected, having been collected at the end of the dry season; those containing some moisture were air-dried. All were processed carefully by hand to remove stones, and were then ground in a large Wiley mill. The ground samples were mixed and subsampled for analysis. After being ground and

mixed, the samples were held at 0°F until they could be subsampled and analyzed.

#### EXTRACTION

Soils—A 100-g subsample was extracted with hexane-acetone (41+59 azeotropic) in a Soxhlet extractor for 16 hr (16).

Forest Floor—A 25-g subsample was extracted with acetone in a Soxhlet extractor for 16 hr.

#### CLEANUP

The soil and forest floor extracts were transferred to individual separatory funnels and water was added to form a water-acetone (2+1) solution. The pesticides were partitioned into hexane by shaking with three 100-ml aliquots of hexane (10).

The hexane extracts were dried with anhydrous sodium sulfate, evaporated to 5-10 ml, and transferred to a 15-g Florisil column (25). The pesticides were eluted from the column with 100 ml dichloromethane—hexane (1+3). Dichloromethane was removed by evaporation, and samples were transferred with hexane to volumetric flasks for analysis.

#### ANALYSIS

The concentrations of DDE, o,p'-DDT, TDE, and p,p'-DDT were 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; TDE, 8.0 minutes; and p,p'-DDT, 9.5 minutes. Other instrument parameters and operating conditions were:

C-1	
Column:	

Pyrex, 180 cm  $\times$  2 mm ID, packed with 5 percent QF-1 (first 126 cm) and 5 percent DC-11 (last 54 cm) on 60-80-mesh Gas Chrom Q, preconditioned for 48 hr at 220°C

Temperatures:

column 185°C detector 190°C injector 205°C

Carrier gas:

Nitrogen flowing at 30 ml/minute

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

	% RECOVERY (RANGE)				
FORM OF DDT	Soil	FOREST FLOOR			
DDE	99 (92–103)	97 (93–100)			
o,p'-DDT	82 (71–99)	99 (96–100)			
TDE	82 (78–91)	85 (80-84)			
p,p'-DDT	97 (92-100)	94 (90–97)			

#### CONFIRMATION

Some industrial pollutants are similar to DDT in structure and properties and can interfere with its detection or identification (12, 14, 17, 19, 20); naturally occurring plant or soil substances may also cause analytical errors (9, 13). To confirm apparent DDT residues in the present study, about half the samples were analyzed by gas-liquid chromatography (GLC) with a chloridespecific, microcoulometric detection system. 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 (PCBs) as DDT isomers and metabolites. Therefore, all samples analyzed with the microcoulometric detector were hydrolyzed with alcoholic potassium hydroxide which would chemically after DDT and TDE, but not PCBs (10). Hydrolyzed samples were then re-analyzed by both electron-capture and microcoulometric detection systems. TDE, o,p'-DDT, and p,p'-DDT peaks disappeared after hydrolysis, indicating that PCBs were not present in detectable quantities and that the quantitative measurement of DDT isomers and metabolites by the electron-capture detection system was correct.

Mass spectrophotometry is the most positive means of identifying pesticides in biological samples, but in the present study, only the forest floor samples and a few of the soil samples contained sufficient DDT to allow use of this technique. Ten forest floor and two soil samples were extracted and purified for analysis. The DDT isomers and metabolites were separated by chromatography of the final hexane extract on 500-μm silica gel H thin-layer plates developed with benzene-hexane (4+96). DDT standards were co-chromatographed on both edges of the 20 imes 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 percent silver nitrate and exposing the plate to ultraviolet light for 15 minutes. The o,p'-DDT, p,p'-DDT, and p,p'-DDE were scraped from the appropriate section of the center of the plate, extracted from the silica gel with hexane, and analyzed by electron-capture gas chromatography. 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 Varian MAT Model CH 7 mass spectrometer with a direct inlet probe. The mass spectra for o,p'-DDT, p,p'-DDT, and p,p'-DDE isolated from the forest floor samples agreed with spectra of appropriate standards and with published spectra (21). A comparison of sample spectra with published PCB spectra (4) showed that no PCBs were present in the isolated pesticides. All confirmation steps gave positive evidence that the isolated and measured substances were indeed

DDT isomers and metabolites and that PCBs were not present in detectable quantities.

## Results and Discussion

# COAST RANGE TRANSECTS

DDT residues were found in all samples, even though all but one site had never received a direct application of insecticide. Mean concentrations of SDDT residues in the forest floor were 0.049 ppm at the coast and 0.047, 0.064, 0.075, and 0.119 ppm at 16, 32, 48, and 64 km inland, respectively, over all four transects (Table 2). These DDT residue levels were quite low and tended to increase progressively from the Oregon coast inland across the coastal mountain range to the western edge of cultivated inland valleys. The two northern transects terminate in forested areas adjacent to the Willamette Valley. No major insect control projects have been conducted in the coastal mountain areas of western Oregon, and the only source of DDT which could have produced these residues appears to be aerial drift from agricultural applications and municipal spray programs for control of mosquitoes and other insect pests.

TABLE 2. \(\Sigma DDT\) residues in forest floors and soils of Coast Range transects of western Oregon,

September-November 1966

		RESIDUES, PPM1					
	C	Dı	STANCE	FROM (	COAST,		Τ
TRANSECT	SAMPLE DEPTH, CM	0	16	32	48	64	TRANSECT MEANS
Wilson	Forest floor	0.052	0.056	0.058	0.058	0.132	0.071
River	Soil: 0-7.5	0.005	0.005	0.006	0.009	0.024	0.010
	Soil: 7.5-15	0.006	0.011	0.013	0.003	0.006	0.008
Yaquina	Forest floor	0.037	0.038	0.081	0.098	0.116	0.074
	Soil: 0-7.5	0.007	0.013	0.006	0.009	0.009	0.009
	Soil: 7.5–15	0.006	0.005	0.005	0.004	0.005	0.005
Umpqua	Forest floor	0.054	0.046	0.060	0.054	0.130	0.069
	Soil: 0-7.5	0.006	0.006	0.008	0.008	0.013	0.008
	Soil: 7.5–15	0.004	0.004	0.005	0.007	0.008	0.005
Rogue	Forest floor	0.054	0.049	0.058	0.093	0.098	0.070
8	Soil: 0-7.5	0.004	0.011	0.007	0.007	0.012	0.008
	Soil: 7.5–15	0.005	0.006	0.005	0.006	0.010	0.006
Site means	Forest floor	0.049	0.047	0.064	0.075	0.119	0.071
	Soil: 0-7.5	0.005	0.009	0.007	0.008	0.015	0.009
	Soil: 7.5–15	0.005	0.006	0.007	0.005	0.007	0.006

NOTE:  $\Sigma$ DDT residues include p,p'-DDE, o,p'-DDT, TDE, and p,p'-DDT. TDE could not be quantified in all samples. <sup>1</sup> Each value represents the mean total residue for four samples from

that site on the transect.

Contamination of the forested areas is surprisingly uniform along the four transects, the highest residue levels occurring adjacent to areas of agricultural activity and population centers in the inland valleys and then decreasing toward the coast with greater distance from the source. Mean annual precipitation varies from 114 cm to 265 cm between the sampling points, but there is no apparent correlation between residue levels and amount of precipitation (r=0.16). Even the lowest

amount of precipitation was apparently adequate to bring down any aerially transported residues to the forest floor.

Residue levels of \(\Sigma\)DDT in samples of the surface layers of mineral soil were much lower than levels found in forest floor samples, with average concentrations of 0.009 ppm and 0.006 ppm in the 0 to 7.5- and 7.5 to 15-cm depths, respectively. Residue levels in the mineral soil showed similar but less pronounced trends in distribution compared to those observed in the forest floor. Residues reaching the forest floor were not readily leached into the mineral soil. More than 80 percent of the \(\Sigma\)DDT residues reaching each site remained in the forest floor.

#### CASCADE RANGE TRANSECTS

Residue levels of  $\Sigma$ DDT in the forest floor and soils at sampling sites along the east-west transects across the Cascade Range in western Oregon appear in Table 3. These sites were selected on the basis of elevation except that for the eastern site on each transect, a location 16 km east of the crest of the passes through the Cascades was selected in order to determine whether the rain shadow had any measurable effect on the level of DDT residues in the forest environment. Residue concentrations in the forest floor were three to five times higher than those found in the Coast Ranges, but were still below 0.50 ppm. Trends in residue distribution patterns are not very distinct, but in general, total residues levels increased with increasing precipitation (r = 0.67) and decreased quite sharply east of the crest.

TABLE 3. \(\simeg DDT\) residues in forest floors and soils of Cascade Range transects of western Oregon,

September-November, 1966

,	- out viz			RESIDU	ES, PPM	1	
		EVATIO	N ABOVE	SEA L	EVEL, N	[ETERS	
Transect	SAMPLE DEPTH, CM	457	762	1067	1372 I	16 км	TRAN- SECT MEANS
Mt. Hood	Forest floor Soil: 0-7.5 Soil: 7.5-15	0.344 0.010 0.006	0.433 0.009 0.007	0.326 0.010 0.005	0.417 0.011 0.006	0.271 0.013 0.010	0.358 0.011 0.007
Santiam Pass	Forest floor Soil: 0-7.5 Soil: 7.5-15	0.330 0.022 0.012	0.268 0.010 0.005	0.391 0.013 0.006	$\begin{array}{c} 1.405^3 \\ 0.081 \\ 0.008 \end{array}$	0.147 0.011 0.006	0.508 0.027 0.007
Willamette Pass	Forest floor Soil: 0-7.5 Soil: 7.5-15	$0.383 \\ 0.010 \\ 0.007$	0.425 0.047 0.010	0.298 0.018 0.008	0.225 0.005 0.004	0.059 0.005 0.003	0.278 0.017 0.006
Crater Lake	Forest floor Soil: 0-7.5 Soil: 7.5-15	0.111 0.013 0.005	0.110 0.012 0.007	0.138 0.007 0.004	0.192 0.007 0.005	0.105 0.008 0.006	0.131 0.009 0.006
Site means	Forest floor Soil: 0-7.5 Soil: 7.5-15	0.292 0.014 0.007	0.309 0.020 0.007	0.288 0.012 0.006	0.560 0.026 0.006	0.145 0.009 0.006	0.319 0.016 0.006

<sup>&</sup>lt;sup>1</sup> Each value represents the mean of four samples at that site.

 <sup>&</sup>lt;sup>2</sup> Eastern sampling point was located 16 km east of the crest of the Cascades.
 <sup>3</sup> This sampling site by chance fell in an old spray unit treated in 1953.

Residue levels in the forest floor samples from the Cascade Range varied considerably between sampling sites along each transect, and even between sublocations at each site. This variation can be attributed largely to old spray projects which took place along the crest of the Cascades from Mt. Hood on the north to Waldo Lake on the south (Fig. 1). Only one sampling site was located in an area that had been sprayed with DDT, but several others were close to an old spray project or were at locations that may have received drift from more than one spray application. Spray projects were conducted along the crest of the Cascades every year from 1949 through 1953, and 1.16 million hectares were treated with a total of 1.30 million kg DDT. None of the treated areas was included in more than one control project, but adjacent units were sprayed in consecutive years. The sampling site at 1,372-meter elevation on the Santiam Pass transect was sprayed in 1953, and 13 years later the forest floor at this site still contained an average concentration of 1.405 ppm **DDT**. The low mean annual temperature at this highelevation site would greatly reduce the rate of normal degradation.

Levels of **\(\Sigma\)DDT** residues in the surface of 7.5-cm layer of mineral soil are almost double the concentration found at the same depth in the Coast Range samples, but the mean concentrations at the 7.5-15-cm depth are identical. The resistance of DDT residues in the forest floor against downward transport by leaching, even at mean annual precipitation levels of 305 cm, is markedly evident in these data.

Mean annual precipitation (Table 4) is lower for each site along the Crater Lake transect relative to the three transects to the north. This transect also crosses the Cascade Range approximately 80 km south of any spray project conducted along the crest of the Cascades. The combined effect of these factors has resulted in a mean SDDT residue level (0.131 ppm) in the forest floor for this transect that is less than half the mean concentration for the other transects. Yet this residue level is considerably higher than the mean concentration of DDT in the forest floor of any of the transects across the Coast Ranges (Table 2). Aerial drift of DDT residues from the large spray projects conducted 80–322 km to the north has contributed to this level of contamination of unsprayed forests.

## DDT ISOMERS AND METABOLITES

Residue levels of each isomer and metabolite of DDT in the forest floor and soil samples show essentially the same relationships and trends along transects of both mountain ranges except for TDE residues in samples from the Coast Ranges. Total residue levels were relatively low in samples from these transects, and the levels of TDE were too low to be quantified in more than

half of the forest floor samples and in almost 75 percent of the soil samples from both depths. In contrast, residues of TDE could be easily quantified in over 75 percent of all samples from the Cascade Range.

TABLE 4. Mean annual precipitation at each sampling site along transects across the Coast and Cascade Ranges in western Oregon, September–November, 1966 <sup>1</sup>

_		PRE	ECIPITATION	N, CM	
		DISTAN	CE FROM (	COAST, KN	1
COAST RANGE — TRANSECTS	0	16	32	48	64
Wilson River	241	254	267	254	127
Yaquina	152	178	203	216	140
Umpqua	165	203	216	152	114
Rogue	203	229	267	216	203
Consum Rower	Eı	EVATION A	BOVE SEA	LEVEL, M	[ETERS
CASCADE RANGE	457	762	1067	1372	16 km F

CASCADE RANGE	EL	EVAIION A	BOVE SEA	LEVEL, IVI	ETERS
TRANSECTS	457	762	1067	1372	16 km E
Mt. Hood	190	203	216	229	127
Santiam Pass	267	279	305	190	89
Willamette Pass	114	152	165	165	76
Crater Lake	102	114	127	152	64

 $^{\rm 1}$  Mean annual precipitation levels were estimated from U.S. Geological Survey isohyetal maps for western Oregon.

The largest proportion of all DDT residues found in samples from both sets of transects was present as p,p'-DDT. Residues of this isomer accounted for approximately 64 percent of the  $\Sigma$ DDT found in the forest floor (Table 5). The proportion of p,p'-DDT decreased with depth, whereas the relative amounts of o,p'-DDT and p,p'-DDE increased with depth. However, more than 80 percent of the total residues found at all sites remained in the forest floor. The relative proportions of the DDT isomers and metabolites, and the changes in relative distribution with sample depth, indicate that degradation is taking place slowly.

TABLE 5. Mean residue levels of DDT isomers and metabolites in forest floors and soils of the Coast and Cascade Range transects expressed as a percentage of the mean \( \Sigma DDT \) residue level, September-November 1966

	-			
SAMPLE DEPTH, CM	p,p'-DDE	o,p'-DDT	p,p'-TDE	p,p'-DDT
	COAST RAN	GE TRANSE	CTS	
Forest floor	14.93	21.35	1	63.72
Soil: 0-7.5	22.38	27.31	_	50.31
Soil: 7.5–15	23.16	35.99	_	40.85
C	CASCADE RA	NGE TRANS	SECTS	
Forest floor	15.82	14.62	4.99	64.57
Soil: 0-7.5	19.70	26.34	4.33	49.63
Soil: 7.5-15	21.48	35.77	2.62	40.13

<sup>&</sup>lt;sup>1</sup> TDE is not included because this metabolite could only be quantified in approximately 35 percent of the litter and soil samples from the Coast Range transects.

SIGNIFICANT EFFECTS OF TRANSECT, SITE, AND SAMPLING DEPTH

Residue data for each range were analyzed statistically to determine whether sampling transects or sampling sites (distance from coast or change in elevation) significantly influenced the level of DDT residue found. A split-split plot design was used, and the data were analyzed by analysis of variance. The first stage examined the influence of subsite and site over all three sampling depths, and the second stage provided a separate analysis for each depth. In addition, residue data for the Cascade Range were analyzed by a factorial analysis of variance using a randomized complete block design.

The average **\(\Sigma\)DDT** residue over all three sampling depths increased significantly with distance inland from the coast (Table 6). Average concentrations over all depths and for all four transects were 20.0, 20.9, 26.0, 29.6, and 46.9 ppm DDT for the sampling sites at 0, 16, 32, 48, and 64 km inland, respectively. Since these means represent 240 individual samples, this upward trend is not likely to be a chance occurrence. Sampling depth and the site X depth interaction were also highly significant (P < 0.01). These results were not unexpected since it was anticipated that if any measurable DDT residues were found, the highest concentrations would occur in the forest floor which represents the receiving surface for any atmospheric residues brought down by precipitation. The site X depth interaction indicates that the upward trend in residues with distance inland occurs at each depth, but the trend is less pronounced at each greater sampling depth.

TABLE 6. Statistical analysis of the influence of transect, site, and subsite over all sampling depths on the distribution of total residues of DDT in the Coast Ranges, September–November 1966

Source	DF	SUM OF SQ.	MEAN SQ.	F-VALUE	
Total	239	300,147			
Transects	3	160	53	n.s.	
Site	4	22,888	5,722	16.16**	
Error (a)	12	4,250	354		
Subsite	3	230	77	n.s.	
Subsite × site	12	601	50	n.s.	
Error (b)	36	3,955	110		
Depth	2	215,228	107,614	286.97**	
Site × depth	8	32,723	4,090	10.91 **	
Subsite × depth	6	483	81	n.s.	
Error (c)	24	9,006	375		
Error (d)	72	7,017	97		

NOTE: \*\* = Differences are highly significant (P < 0.01); n.s. = differences are not significant (P > 0.05).

Analysis for each depth did not provide any additional information. The influence of site showed the same highly significant increase with distance inland for forest floor residues. This relationship was only significant (P < 0.05) for residues in the surface layer of mineral soil, and nonsignificant (P < 0.05) for residue levels in the lower soil samples.

A similar series of analyses was conducted for each DDT isomer and metabolite. Residue levels of p,p'-

DDE, o,p'-DDT, and p,p'-DDT showed the same relationships discussed above and the same levels of significance.

Residue data for the Cascade Range transects were statistically analyzed by the same split-split plot design (Table 7). There were no significant differences among average SDDT levels by transect or by sites for a given transect for either forest floor or soil. However, examination of the data presented in Table 3 indicates that residue levels are generally lower at each site located 16 km east of the crest of the Cascades, and that residue levels along the Crater Lake transect are lower than those observed for each of the three transects crossing the Cascade Range closer to the old spray projects. Residue data for the forest floor samples were analyzed by a factorial analysis of variance using a randomized complete block design. Residue levels in the forest floor were significantly lower east of the Cascades, and residues along the Crater Lake transect were significantly lower than residue levels in the forest floor samples from the other three transects (both at P < 0.01).

TABLE 7. Statistical analysis of the influence of transect, site, and subsite over all sampling depths on the distribution of \(\SigmaDDT\) residues in samples from the Cascade Range, September–November 1966

Source	DF	SUM OF SQ.	MEAN SQ.	F-VALUE
Total	239	15,684,060		
Transects	3	542,593	180,864	1.81 n.s.
Site	4	517,790	129,448	1.29 n.s.
Error (a)	12	1,198,556	99,880	
Subsite	3	103,352	34,451	1.22 n.s.
Subsite × side	12	290,760	24,230	0.86 n.s.
Error (b)	36	1,012,599	28,128	
Depth	2	5,048,995	2,524,498	30.93**
Site × depth	8	922,449	115,306	4.69**
Subsite × depth	6	183,554	30,592	1.24 n.s.
Error (c)	24	1,958,987	81,624	
Error (d)	72	1,771,308	24,601	

NOTE: \*\* = Differences are highly significant (P < 0.01); n.s. = differences are not significant (P > 0.05).

This distribution of DDT residues in unsprayed areas is consistent with similar data reported by Cory et al. (7). In a study of the distribution of DDT residues in the Sierra Nevada Mountains, concentrations were markedly lower east of the crest. They also reported higher and more variable residue levels in samples from locations near old spray projects conducted in 1953 and 1956.

Closer examination of the data for each subsite along the Cascade Range transects shows that residue levels from all sampling sites and depths were extremely variable. The standard deviation of the mean for SDDT residues in the forest floor samples from the Santiam transect is greater than the mean. The causes of this variation are most likely the relative position of the sampling sites in reference to the location of the old

spray projects and the prevailing climatic conditions at the time those 1.16 million hectares were sprayed with DDT. The one sampling site that unintentionally fell in an old spray unit greatly distorts the residue levels for that site and for the transect, but it does not account for all variation observed among and between sampling sites. This discussion applies equally to all statistical analyses conducted on the Cascade Range residue data for each depth and for each DDT isomer and metabolite.

#### Conclusions

The primary purposes of this study were to determine to what extent untreated forested areas in western Oregon may have become contaminated through washout of DDT residues known to be present in the atmosphere, and to establish background levels of DDT residues in forest litter and soil. Perhaps the most important result obtained is the fact that measurable quantities of DDT were found at every site sampled along each of the eight transects. Residue levels along transects across the Coast Ranges increased significantly with distance inland from the coast. This trend is readily explained in that each transect ends in a forested area adjacent to agriculturally important inland valleys or population centers which serve as a source of the pesticide residues. Residue levels along transects across the Cascade Range were considerably higher than those found in the Coast Ranges because of the influence of a number of large insect control projects conducted along the crest of the Cascades between 1949 and 1953. Distance and direction of sampling sites from these sprayed areas have produced a wide variation in residue levels which tends to mask the influence of annual rainfall distribution and other local differences in topography.

Although DDT residues were found at every point sampled, the levels of these residues were generally low. Maximum concentrations of DDT in forest floor samples from the Coast Ranges did not exceed 0.17 ppm. Residue levels in forest floor samples from the Cascade transects were generally three to five times higher than those from Coast Ranges, but mean residues still did not exceed 0.50 ppm DDT. Low levels in the surface mineral soils of both sets of transects further substantiate the fact that DDT is not readily leached into forest soils. The tenacity with which the organic horizons of forest soils hold residues of DDT strongly suggests that the levels of residue measured are not likely to become available to nontarget organisms in toxic amounts (28).

#### LITERATURE CITED

 Abbott, D. C., R. B. Harrison, J. O'G. Tatton, and J. Thomson. 1965. Organochlorine pesticides in the atmospheric environment. Nature 208(5017):1317-1318.

- (2) Acree, F., Jr., M. Beroza, and M. C. Bowman. 1963. Codistillation of DDT with water. J. Agric. Food Chem. 11(4):278-280.
- (3) Antommaria, P., M. Corn, and L. DeMaio. 1965. Airborne particulates in Pittsburgh: Association with p,p'-DDT. Science 150(3702):1476-1477.
- (4) Bagley, G. E., W. L. Reichel, and E. Cromartie. 1970. Identification of polychlorinated biphenyls in two bald eagles by combined gas-liquid chromatography-mass spectrometry. J. Assoc. Off. Anal. Chem. 53(2):251-261.
- (5) Belyea, G. Y. 1967. A study of DDT residues in soils and a related food chain in northern Maine forests. M.S. thesis, University of Maine, 80 pp.
- (6) Cole, H., D. Barry, and D. E. H. Frear. 1967. DDT levels in fish, streams, stream sediments, and soil before and after DDT aerial spray application for fall cankerworm in northern Pennsylvania. Bull. Environ. Contam. Toxicol. 2(3):127-146.
- (7) Corey, L., P. Fjeld, and W. Serat. 1970. Distribution patterns of DDT residues in the Sierra Nevada Mountains. Pestic. Monit. J. 3(4):204-211.
- (8) Decker, G. C., C. J. Weinman, and J. M. Bann. 1950. A preliminary report on the rate of insecticide residue loss from treated plants. J. Econ. Entomol. 43(6):919.
- (9) Frazier, B. E., G. Chesters, and G. B. Lee. 1970. "Apparent" organochlorine insecticide contents of soils sampled in 1910. Pestic. Monit. J. 4(2):67-70.
- (10) Hamence, J. H., P. S. Hall, and D. J. Caverly. 1965. The identification and determination of chlorinated pesticide residues. Analyst 90(1076):649-656.
- (11) Harris, C. R., and E. P. Lichtenstein. 1961. Factors affecting the volatilization of insecticidal residues from soils. J. Econ. Entomol. 54(5):1038-1045.
- (12) Holmes, D. C., J. H. Simmons, and J. O'G. Tatton. 1967. Chlorinated hydrocarbons in British wildlife. Nature 216(5112):227–229.
- (13) Hylin, J. W., R. E. Spenger, and F. A. Gunther. 1969. Potential interferences in certain pesticide residue analyses from organochlorine compounds occurring naturally in plants. Residue Rev. 26:127–138.
- (14) Koeman, J. H., M. C. ten Noever de Brauw, and R. H. de Vos. 1969. Chlorinated biphenyls in fish, mussels, and birds from the River Rhine and The Netherlands coastal area. Nature 221(5186):1126-1128.
- (15) Middleton, J. T. 1965. The presence, persistence, and removal of pesticides in air. Pages 191-197 in Research in Pesticides. C. O. Chichester (Ed.). Academic Press, New York, N.Y.
- (16) Pionke, H. B., G. Chesters, and D. E. Armstrong. 1968. Extraction of chlorinated hydrocarbon insecticides from soils. Agron. J. 60(3):289-292.
- (17) Reynolds, L. M. 1969. Polychlorobiphenyls (PCB's) and their interference with pesticide residue analysis. Bull. Environ. Contam. Toxicol. 4(3):128-143.
- (18) Riekerk, H., and S. P. Gessel. 1968. The movement of DDT in forest soil solutions. Soil. Sci. Soc. Am. Proc. 32(4):595–596.
- (19) Risebrough, R. W., D. B. Peakall, S. G. Herman, M. N. Kirven, and P. Rieche. 1968. Polychlorinated biphenyls in the global ecosystem. Nature 220(5172): 1098-1102.
- (20) Risebrough, R. W., P. Reiche, and H. S. Olcott. 1969. Current progress in the determination of polychlorinated biphenyls. Bull. Environ. Contam. Toxicol. 4(4): 192-201.
- (21) Sphon, J. A., and J. N. Damico. 1970. The mass spectra of some chlorinated aromatic pesticidal compounds. Org. Mass Spectrom. 3(1):51-62.

- (22) Tabor, E. 1965. Pesticides in urban atmospheres. Paper No. 65-30 presented at the 58th Annual Meeting of the Air Pollution Control Association, Toronto, Canada, June 20-24, 1965.
- (23) Tarrant, R. F., D. G. Moore, W. B. Bollen, and B. R. Loper. 1972. DDT residues in forest floor and soil after aerial spraying, Oregon—1965-68. Pestic. Monit. J. 6(1):65-72.
- (24) Wheatley, G. A., and J. A. Hardman. 1965. Indications of the presence of organochlorine insecticides in
- rainwater in central England. Nature 207(4996):486-487
- (25) Wood, B. J. 1966. Elution of dieldrin and endrin from Florisil. J. Assoc. Off. Anal. Chem. 49(2):472-473.
- (26) Woodwell, G. M. 1961. Persistence of DDT in a forest soil. For. Sci. 7(3):194-196.
- (27) Woodwell, G. M., and F. T. Martin. 1964. Persistence of DDT in soils of heavily sprayed forest stands. Science 145(3631):481–483.
- (28) Yule, W. N. 1970. DDT residues in forest soils. Bull. Environ. Contam. Toxicol. 5(2):139-143.

Reproduced from Pesticides Monitoring Journal, Vol. 14, No. 3, December 1980, by the FOREST SERVICE, U.S. Department of Agriculture, for official use.

4---