

DDT Levels in Fish, Streams, Stream Sediments, and Soil Before and After DDT Aerial Spray Application for Fall Cankerworm in Northern Pennsylvania¹

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The Pennsylvania Department of Forests and Waters used DDT early in 1965 to suppress an extensive outbreak of the fall cankerworm (Alsophila pomataria) in northern hardwood stands in Potter and Tioga counties. Aerial application was made in May 1965 to approximately 104,000 acres of forest land.

The area treated consisted of maple, beech, birch, cherry, and oak in mixed stands growing in terrain characterized by high ridges and narrow valleys interspersed with small streams containing native brook trout (Salvelinus fontinalis) and varying populations of white suckers (Catostomus commersoni).

The area is accessible only by a few unpaved roads and contains almost no habitation, being over 90 percent state forest land. There were no previous records of DDT or other chlorinated hydrocarbon pesticide used in the area and much of it was seldom visited by humans, except for occasional hunters and fisherman.

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Because of the absence of previous pesticide use it was felt that this area presented an ideal opportunity to evaluate the accumulation of a single application of DDT in certain phases of the ecosystem.

Locations

Two streams and their watersheds were selected for study: Sunken Branch and Lyman Run. These two streams located in the center of the treatment area ultimately flow into the west branch of Pine Creek and thence into the west branch of the Susquehanna River. Lyman Run, which contains an appreciable population of white suckers is blocked by a high (approximately 20 ft.) dam in its middle reaches which prevents upstream migration of these fish. Thus suckers present in the headwaters area represent a relatively stable nonmigratory population. Sunken Branch, a smaller cold water stream, contained no suckers. In addition it is fed to a greater degree by underground sources and contains much less bottom sediment and silt deposits. Both streams contained native brook trout.

Sample Collection Methods

In the week immediately before any spray treatments were applied, native brook trout, white suckers (Lyman Run only), crayfish, water, stream sediments, and watershed soil samples were collected to determine if residues of any persistent chlorinated hydrocarbon pesticides were present. Collection methods were as follows:

1. Native brook trout and suckers: Fish were collected with electrofishing gear with a portable gasoline 230 volt generator. On Lyman Run two collection stations were established, one immediately below the confluence of the small headwater tributaries (hereafter named splash dam site) and another area approximately 1 mile downstream from the first site (hereafter named ECC112 site). At each site both suckers and brook trout were collected from a stretch of water approximately 100 yards long. Ten brook trout and 10 suckers from 6 to 10 (15-25 cm) inches long were selected for analysis at each collection station. On Sunken Branch, the smaller stream, one collection station was established about 1/4 mile below the confluence of its tributaries. Again 10 trout 6 to 10 inches long were collected for analysis. All samples were stored in glass jars under refrigeration and were extracted for analysis within 48 hours of collection.
2. Stream Sediments: In Lyman Run 4 composite sediment samples were taken. Subsamples were collected and composited from each of the 100-yard stretches of stream where the fish were collected and another sample was composited from the 100 yards of the stream immediately above each fish collection station. Thus on Lyman Run four samples representing 400 yards of stream were obtained. In Sunken Branch two samples representing 200 yards of stream were taken. The samples taken were composed of organic matter, silt, and clay fractions which had accumulated in the pool bottoms and quieter areas of the

streams. A small hand scoop was used to gather the subsamples of bottom sediment. Each sample was a composite of 20 separate collections from different portions of the stream and in all instances represented the top half inch (1 cm) of sediment. Deeper muds were not sampled. The final volume of the composite sediment sample was one quart. All samples were stored in glass jars under refrigeration and were extracted for analysis within 7 days of collection.

3. Water: Two water samples of four gallons each were collected from Lyman Run before spray treatment. These were collected in one-gallon jugs.
4. Crayfish: These were collected in the same stream areas as the fish samples and were obtained by hand picking or from a drift net after dislodging stones upstream. In Lyman Run two samples of 10 specimens each were obtained. On Sunken Branch crayfish were much scarcer and a sample of four specimens was obtained with difficulty. All samples were stored in glass jars under refrigeration and were extracted for analysis within 48 hours of collection.
5. Watershed soil samples: Two composite samples of soil were collected from the watershed area of Lyman Run and two composite samples from the Sunken Branch area. Each composite sample was composed of 20 subsample collections from different random sites and covered an area ranging from the banks of the streams to several hundred feet up on the ridges. In sampling a profile consisting of the forest litter (leaves, debris, and

decomposed organic matter) plus the top inch of mineral soil beneath the litter was taken. The final weight of each composite sample was approximately five pounds. All samples were refrigerated in polyethylene bags and were extracted within two weeks of collection.

DDT Spray Treatment

Aerial spray applications were made between May 10 and May 31, 1965, with a total of 104,560 acres sprayed by the end of the operation. Spraying of the residue study area was done between May 20 and May 25. The DDT was formulated at the rate of 0.5 pound of technical DDT in 0.6 quart of naphtha plus sufficient No. 2 fuel oil to make 1 gallon of solution. One gallon of this 8 percent DDT solution, containing 0.5 pound of technical DDT, was applied per acre.

Application was made in spray swaths 400 feet wide at the release rate of 125 gallons of solution per minute at 165 miles per hour and 75 to 100 feet above the tree tops.

Post-Treatment Sample Collection

Water samples were collected from the same Lyman Run site as the pre-treatment samples. The first was taken one hour after spraying the headwaters area of Lyman Run and a second sample was taken 14 days later. The first post-treatment sample collections of other materials were made 32 days after completion of the entire spray application. Fish, crayfish, and stream sediments were collected in the same manner and quantities from the same sites as the pre-treatment collections. One hundred twenty-two days

(approximately four months) after treatment another series of samples was collected. The final collections were made 380 days after treatment. This included extensive watershed soil sampling involving 10 composite samples (200 individual subsamplings) from the Sunken Branch watershed and 10 from the Lyman Run watershed as well as 5 composite samples from an untreated area approximately 30 miles away.

Because of extensive leaf cover in the treated area post-treatment watershed soil sample collection was delayed until one year following treatment when all treated deciduous foliage had reached the forest floor and wash-off from tree branch and twig surfaces had occurred.

As a check, sediment and soil samples on Young Woman's Creek and watershed (an untreated area 30 miles away) were collected. This collection included five composite soil samples and two composite stream sediment samples. These were taken on the same date as the 380 day post-treatment samples from Lyman Run and Sunken Branch.

Sample Extraction and Cleanup Procedures

Fish and Crayfish. A weighed sample of approximately 200 grams was macerated in a Waring Blendor with 300 ml. of chloroform-methanol (2:1) and approximately 100 grams of anhydrous sodium sulfate added during the blending process. The liquid was decanted, and the blending repeated with another 300-ml. portion of chloroform-methanol. The extracts were combined and filtered with suction through filter paper on a Büchner funnel. The

filtered extract was then placed in a flask equipped with a Snyder column. This was placed on the steam bath and the chloroform removed. One hundred ml. of n-hexane was added, the liquid transferred to a separatory funnel and washed thoroughly three times with 200-ml. portions of water to remove the methanol. The washed extract was evaporated to a small volume in a flask on the steam bath, then taken to dryness with a stream of air at room temperature.

At this point the residue consisted of lipid material in a semisolid state. Two grams of this were weighed into a small separatory funnel and dissolved in 25 ml. of petroleum ether. This solution was extracted by shaking for one minute with 25 ml. of acetonitrile saturated with petroleum ether; the acetonitrile layer was drawn off and the lipid solution re-extracted with three additional 25-ml. portions of acetonitrile saturated with petroleum ether. The combined acetonitrile extracts were evaporated to a small volume and taken up in n-hexane. This was then evaporated in a Kuderna-Danish evaporator to exactly 2 ml. and an aliquot injected into the gas chromatograph.

Stream Sediments and Soil. One-hundred-gram samples of moist sediment or soil were placed in large Soxhlet extractors and extracted 16 hours with 600 ml. of a n-hexane-acetone (2:1) mixture. The extract was washed with water in a separatory funnel to remove the acetone, then passed through a column of alumina, Celite and Nuchar activated carbon (2:1:1). This removed any pigments and other interfering substances. The purified extract was

concentrated in a Kuderna-Danish evaporator to a volume of 2 ml. and an aliquot injected into the gas chromatograph.

Water. Four-liter samples of water were exhaustively extracted in a Pope liquid-liquid extractor with n-hexane. The extracts were concentrated to 0.2 ml. in a Kuderna-Danish apparatus heated by a steam bath.

Analytical Procedure

All analyses were made on a Research Specialties Gas Chromatograph Model 600, equipped with a 6-foot column packed with Gas-chrome Q impregnated with 5 percent DC-200. The column temperature was maintained at 210° C., the detector at 270° C., with a nitrogen flow of 60 ml. per minute. Samples of standard solutions were run periodically to check on recovery. All results were calculated on a fresh or "as received" basis. Considering the size of sample and analytical method, the level of confidence of the results on the water samples was 0.0001 p.p.m., and on all other samples 0.002 p.p.m. In some instances soil and sediment residue traces less than 0.002 p.p.m. were found. These were reported as NR (no residue) because these values were not statistically significant when checked for reproducibility in split replicated samples.

Results

Pre-treatment Sampling. Analysis of pre-treatment watershed soils in both Sunken Branch and Lyman Run areas indicated the presence of small quantities of DDE, p,p'-DDT and dieldrin. Levels ranged from 0.002 to 0.005 p.p.m. for dieldrin, from 0.002 p.p.m. to 0.004 p.p.m. for DDE and from 0.005 to 0.009 p.p.m. for p,p'-DDT

(Table 1). Most of these were near the analytical limit of confidence. No other persistent chlorinated pesticides were detectable.

Water from Lyman Run before any treatment in the area contained no detectable residues. However, after treatment started in the vicinity, but before actual spraying of the Lyman Run watershed, minute quantities of p,p' -DDT (0.0052 p.p.m.) were detected in the water (Table 3).

Stream bottom sediments in Lyman Run and Sunken Branch contained DDE, and p,p' -DDT in amounts ranging from no detectable residue (less than 0.002 p.p.m.) to 0.005 p.p.m. for DDE, and from 0.008 p.p.m. to 0.012 p.p.m. for p,p' -DDT. In addition, Lyman Run sediments from one site contained 0.003 p.p.m. of dieldrin (Table 2). Sunken Branch sediments did not contain any dieldrin.

Brook trout contained DDE, TDE (DDD), p,p' -DDT and dieldrin in quantities ranging up to 0.42 p.p.m. for DDE, 0.10 p.p.m. for TDE, 0.54 p.p.m. for p,p' -DDT, and 0.11 p.p.m. for dieldrin. Sunken Branch brook trout did not contain any dieldrin. White suckers in Lyman Run contained DDE, p,p' -DDT and dieldrin at levels up to 2.40 p.p.m. for DDE, 3.7 p.p.m. for p,p' -DDT, and 1.8 p.p.m. for dieldrin (Table 4).

Crayfish contained DDE, and p,p' -DDT at levels up to 1.1 p.p.m. for DDE and 1.90 p.p.m. for p,p' -DDT. No crayfish contained dieldrin (Table 4).

Post-treatment Sampling. A water sample collected 1 hour after spraying the headwaters area contained 0.0240 p.p.m.

TABLE 1

Residues in Watershed Soils Before and After Aerial
Application of 0.5 lb. of DDT per Acre (P.P.M.¹).

Location	Pre-treatment, May 1965			
	DDE	<u>o,p'</u> -DDT	<u>p,p'</u> -DDT	Dieldrin
SUNKEN BRANCH AREA				
East slope	0.003	NR ²	0.006	0.002
West slope	0.004	NR	0.005	0.003
LYMAN RUN AREA				
East slope	0.002	NR	0.009	0.005
West slope	0.004	NR	0.007	0.003
Post-treatment, May 1966 ^{3,4}				
SUNKEN BRANCH AREA				
East slope	0.025	0.120	0.220	0.003
West slope	0.038	0.118	0.270	0.003
LYMAN RUN AREA				
East slope	0.004	0.042	0.138	0.006
West slope	0.008	0.072	0.018	0.004
YOUNG WOMAN'S CREEK				
AREA ⁵	0.003	0.006	0.009	NR

¹ Based on soil weight as collected.

² NR = No detectable residue (less than 0.002 p.p.m.).

³ 380 days following application.

TABLE 2

Residues in Stream Bottom Sediments Before and After Aerial
Application of 0.5 lb. of DDT per Acre (P.P.M.¹).

Location	DDE	<u>o,p'</u> -DDT	<u>p,p'</u> -DDT	Dieldrin
SUNKEN BRANCH				
Pre-treatment	0.002 ²	NR ³	0.012	NR
32 d/treatment	0.002	0.003	0.003	NR
122 d/treatment	NR	NR	0.002	NR
380 d/treatment	0.003	NR	0.002	NR
LYMAN ECC112 site				
Pre-treatment	0.005	NR	0.008	NR
32 d/treatment	NR	NR	0.002	NR
122 d/treatment	0.002	NR	0.008	NR
380 d/treatment	0.004	NR	NR	NR
LYMAN RUN Splash dam site				
Pre-treatment	0.002	NR	0.009	0.003
32 d/treatment	NR	0.003	0.009	NR
122 d/treatment	0.002	NR	NR	NR
380 d/treatment	NR	NR	NR	NR
YOUNG WOMAN'S CREEK ⁵				
No treatment	NR	NR	0.006	NR

¹ Based on wet weight as collected.

² Residue levels reported are mean levels calculated from analytical results of two composite samples one collected from same part of stream as fish sample and one collected from 100 yd. area immediately upstream from fish sample area.

³ NR = No detectable residue (less than 0.002 p.p.m.).

⁴ d/treatment (days post-treatment).

⁵ Samples collected from untreated area at same date as "380 days post-treatment" samples were from spray area.

TABLE 3

Residues in Lyman Run Water Samples Before and After Aerial
Application of 0.5 lb. of DDT per Acre Splash Dam Site.

<u>Sampling time</u>	<u>p,p'-DDT</u>
A. Before any spray treatment in area	less than 0.0001 p.p.m.
B. Before spray treatment of Lyman Run Watershed ¹	0.0052 p.p.m.
C. One hour after spray treatment of Lyman Run Watershed	0.0240 p.p.m.
D. Fourteen days after termination of complete spray program	trace (0.00012 p.p.m.)

¹ Five days of spraying occurred in the vicinity surrounding
Lyman Run Watershed before this sample was collected.

TABLE 4

Residues in Fish and Crayfish in P.P.M.¹ Before and at Varying Intervals After
Aerial Application of 0.5 lb. DDT per Acre.

Site and Time	Brook Trout					White Suckers					Crayfish				
	DDE	TDE	$\frac{0,P'}{DDT}$	$\frac{P,P'}{DDT}$	Diel-drin	DDE	TDE	$\frac{0,P'}{DDT}$	$\frac{P,P'}{DDT}$	Diel-drin	DDE	TDE	$\frac{0,P'}{DDT}$	$\frac{P,P'}{DDT}$	Diel-drin
Sunken Branch															
Pre-treatment	0.07	0.04	NR ²	0.04	NR	None present in Sunken Branch					NR	NR	NR	0.14	NR
32 d/treatment	4.8	4.9	NR	9.1	NR						0.80	0.71	NR	0.92	NR
122 d/treatment	0.03	0.02	NR	0.03	NR						0.05	0.09	NR	0.11	NR
380 d/treatment	0.28	0.06	NR	0.04	NR						0.07	NR	NR	0.09	NR
Lyman Run															
ECC112															
Pre-treatment	0.34	NR	NR	0.54	0.11	2.4	NR	NR	3.7	1.8	NR	NR	NR	0.47	NR
32 d/treatment	4.8	6.1	NR	9.5	NR	4.7	5.7	NR	6.9	NR	0.88	0.56	NR	0.88	NR
122 d/treatment	0.29	0.16	0.12	0.20	NR	0.27	0.13	NR	0.22	NR	0.09	0.04	NR	0.10	NR
380 d/treatment	0.33	0.10	NR	0.23	NR	0.18	0.24	NR	0.24	NR	0.02	NR	NR	0.06	NR
Splash Dam															
Pre-treatment	0.42	0.10	NR	0.37	0.08	0.72	NR	NR	1.7	0.9	1.1	NR	NR	1.9	NR
32 d/treatment	1.5	6.1	NR	10.6	NR	5.0	7.8	NR	6.8	NR	1.4	1.6	NR	1.1	NR
122 d/treatment	0.31	0.11	0.04	0.07	NR	0.83	0.54	0.04	0.51	NR	0.02	0.01	NR	0.01	NR
380 d/treatment	0.36	0.10	NR	0.09	NR	0.25	0.32	NR	0.32	NR	0.06	NR	NR	0.07	NR

¹ Based on fresh weight of whole fish or crayfish. Each sample analysis reported is a composite of 10 fish in the 6-10 inch category.

² NR = No detectable residue (less than 0.002 p.p.m.).

p,p'-DDT. This dropped to 0.00012 p.p.m. 14 days following treatment.

Levels of insecticides in stream bottom sediment samples 32 days after treatment were quite similar to pre-treatment levels and DDE and p,p'-DDT were found at levels up to 0.002 p.p.m. for DDE and up to 0.009 p.p.m. for p,p'-DDT. The only significant change from pre-treatment levels was the finding of 0.003 p.p.m. o,p'-DDT, one of the isomeric forms of DDT commonly present in technical DDT as currently manufactured. Dieldrin was present in quantities less than 0.002 p.p.m.

Brook trout samples 32 days after treatment contained up to 4.80 p.p.m. DDE, 6.1 p.p.m. TDE, 0.12 p.p.m. o,p'-DDT, and 10.6 p.p.m. p,p'-DDT. Dieldrin was not detectable. White suckers in Lyman Run contained up to 5.0 p.p.m. DDE, 7.8 p.p.m. TDE, and 6.9 p.p.m. p,p'-DDT. Dieldrin and o,p'-DDT were not detectable (Table 4).

Crayfish contained up to 1.4 p.p.m. DDE, 1.6 p.p.m. TDE and 1.1 p.p.m. p,p'-DDT. Dieldrin and o,p'-DDT were not detectable (Table 4).

Thus brook trout and white suckers accumulated significant amounts of DDE, TDE, and p,p'-DDT levels in the one-month period following spray treatment. Residues in brook trout increased more than tenfold in some instances, and in white suckers the amounts more than doubled in all instances (Table 4). Amounts in crayfish increased somewhat.

At the end of 122 days following treatment stream sediment levels continued to remain essentially the same as in the pre-treatment sampling. Residues in fish and crayfish returned to pre-treatment levels with the brook trout containing up to 0.31 p.p.m. DDE, 0.16 p.p.m. TDE, 0.12 p.p.m. o,p'-DDT, and 0.20 p.p.m. p,p'-DDT; the white suckers dropped below pre-treatment levels, containing 0.83 p.p.m. DDE, 0.54 p.p.m. TDE, 0.04 p.p.m. o,p'-DDT, and 0.51 p.p.m. p,p'-DDT. Dieldrin was not present. Residues in crayfish also dropped to pre-treatment levels.

At the end of a one-year (380 day) period stream sediment residues were similar to pre-treatment levels with up to 0.004 p.p.m. DDE and 0.002 p.p.m. p,p'-DDT. Dieldrin was present in trace quantities less than 0.002 p.p.m. Traces of o,p'-DDT (less than 0.002 p.p.m.) were also present. This material appeared in the first series of post-treatment samples and declined steadily with time.

The trout and suckers contained approximately the same amounts of pesticides 380 days after treatment as they did 122 days after treatment. The residues in trout were almost exactly the same as before treatment, while the suckers again showed residues lower than in pre-treatment samples.

Watershed soils after one year still contained significant quantities of DDE, o,p'-DDT, p,p'-DDT and traces of dieldrin; for the Sunken Branch area the amounts found ranged from 0.025 to 0.038 p.p.m. for DDE, 0.118 to 0.120 p.p.m. for o,p'-DDT, and 0.220 to 0.270 p.p.m. for p,p'-DDT. Dieldrin remained at very

low levels as before treatment.

Lyman Run area soil samples contained from 0.004 p.p.m. to 0.008 p.p.m. DDE, 0.042 p.p.m. to 0.072 p.p.m. o,p'-DDT, and from 0.018 p.p.m. to 0.138 p.p.m. p,p'-DDT. Dieldrin remained at levels approximately the same as pre-treatment samples (Table 1). Young Woman's Creek soil samples were quite similar to the Lyman Run and Sunken Branch pre-treatment samples.

Discussion and Results

Pre-treatment. The presence of DDE, p,p'-DDT and dieldrin in trace quantities in watershed soils and stream sediments and in somewhat larger quantities in fish and crayfish prior to any known treatment of the area with these materials supports the idea previously expressed by the authors (3) that aerial transport may be an important mechanism of long distance pesticide transfer. Previously, various unexplainable pesticide occurrences have been reported including grains and forages in Pennsylvania by Cole, Barry, and Frear (3), in antarctic seals and penguins, by George and Frear (6) and in rain water in England by Wheatley and Hardman (10).

In addition, Antommaria, Corn, and De Maio (1) reported the occurrence of p,p'-DDT on airborne particulates in the air over Pittsburgh with some of the particulates in size categories that would have little or no sedimentation rates.

In agreement with observations of Hunt and Keith (7) typical biologic concentration occurred within the ecosystem. For example, residues in watershed soils and stream sediment were quite similar, generally occurring at the few parts per billion level while

residues in the trout were approximately twenty to one hundred times higher than stream sediments and residues in white suckers were relatively high-approximately six to fifteen times higher than amounts found in the trout. The most noteworthy example of concentration was in the case of dieldrin where a 0.003 p.p.m. maximum was found in Lyman Run stream sediments, while the white suckers in this same stream contained 1.80 p.p.m.; a 600-fold increase.

TDE was present in both suckers and trout although none was present in watershed soils and stream sediments. This may be explained on the basis of metabolic conversion of p,p'-DDT to TDE by the fish in a manner similar to that reported to occur in rat livers by Datta et al. (5) and in fish by Bridges et al. (2).

Post-treatment. As would be anticipated, the levels of DDE, TDE and p,p'-DDT in fish increased considerably in the month following treatment. However, residue levels in stream sediments did not increase correspondingly but remained at pre-treatment levels during the year following treatment. Residues in trout also returned to pre-treatment values after four months, and one year later were the same as before treatment. These results would indicate that the increase in residues in the fish was due to the immediate effects of the spraying. The increase apparently came about as a result of a small quantity of residue in the water or by the ingestion of insects killed by DDT or both. It is of interest that although the level of p,p'-DDT in the water reached 24 parts per billion (0.024 p.p.m) one hour after spraying at the splash dam site on Lyman Run no observable fish kills were detected

in the study areas of Lyman Run or Sunken Branch. This level is in excess of the LC_{50} for p,p' -DDT reported by various researchers and further illustrates the difficulty in utilizing data on residues in water as pointed out by Marking (8) in his toxicant studies with DDT. In the present instance it is not known how long a significant DDT concentration was maintained in the water at the sampling site. Water sampling at 24 hour intervals or less would have been needed to evaluate the importance of water residues precisely.

The residue levels in white suckers, which were lower in samples taken 122 and 380 days after treatment than in pre-treatment samples are difficult to explain. It is possible that some of the original suckers present in Lyman Run before treatment died or were caught by predators immediately following the spray treatment, so that the later samples represented a different population than the pre-treatment samples. It is also possible that in such small samples the averages could be affected by the presence of a few individuals with abnormally high or low residue levels.

It is of interest that no o,p' -DDT was detectable in the area prior to the spray operation but that after spraying o,p' -DDT was detectable in the treated area watershed soils and stream sediments and in an untreated site 30 miles away. It is unfortunate that this site 30 miles distant on Young Woman's Creek watershed was not sampled in the pre-treatment collections when base lines were established. Hence, it can not be said with complete certainty if the o,p' -DDT traces in the Young Woman's Creek watershed were drift

from the treatment operation.

The absence of dieldrin from the fish following DDT treatment may represent a situation similar to that reported by Street (9) who found that in rats dieldrin storage was depressed by ingestion of DDT. Levels of dieldrin in soil and stream sediments did not change significantly throughout the year.

The results in the present investigation contain both similarities and differences when compared with results reported in prior researches. Cope (4) reported a decrease with time after treatment of DDT and DDE levels in trout. However, the time interval required for the decrease was quite long and before-treatment residue levels in the trout were not available for comparison. Bridges et al. (2) reported that following DDT treatment of a small pond DDT levels in fish rose and then declined slowly. The crayfish levels were about one-half those found in fish. In the present study residues in crayfish were also generally less than residues in fish. Residues in fish in the present study declined much more rapidly than reported in either the Cope (4) or Bridges (2) investigations.

Evaluating the effects of the single application of 0.5 pound per acre it seems that the only long-term change noted was the accumulation of a significant portion of the DDT application in the forest litter and surface soil of the treated area. If these materials are reaching the surface waters draining the area they did not result in a measurable increase in residues in either fish, crayfish, or stream bottom sediments one year later.

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