

# CHLORINATED HYDROCARBONS DEPOSITED IN BIOLOGICAL MATERIAL<sup>1</sup>

## III. SOIL AND LAKES

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Soil may become contaminated with chlorinated hydrocarbons either by direct addition to control a pest harbored in the soil or by chance contamination when crops are treated.

The persistence of DDT in soil was investigated by Fleming and Maines (23). The average per cent of insecticide retained by soil after an initial treatment of 25 lb per acre was as follows: after 1 year, 97%; 2 years, 90%; 3 years, 79%; 4 years, 64%; 6 years, 56%; and 8 years, 44%. The insecticide was most persistent in sand and least in muck. The pH within a range of 4.0 to 7.5 appeared to have no effect on DDT persistence in soil. Soil under apple trees which had been sprayed with DDT for 7 years contained an average of 62.2 lb insecticide per acre while soil from between trees contained 35.5 lb per acre (32).

Other studies on DDT residues in orchard soils were conducted by Chisholm, *et al.* (12). They tested soils from apple orchards located in Indiana, New Jersey and Washington. Orchards in Indiana were sprayed for 5 years prior to the tests. Debris on the surface of soil contained from 96.8 to 176.8 ppm insecticide while the upper 3 inches of soil contained 13.0 ppm and the next 3 inches had 5.6 ppm. New Jersey orchards were sprayed from 2 to 4 years. Residues of DDT in the upper 3 inches of soil ranged from 23.5 to 47.0 ppm and in the next 3 inches from 5.0 to 9.0 ppm. The upper 3 inches of soils from Washington orchards, sprayed for 3 years, contained from 4.0 to 90.6 ppm DDT.

Soils from peach orchards in New Jersey and New York were tested. New Jersey orchards sprayed for 2 or 3 years contained from 18.0 to 37.5 ppm DDT in the upper 3 inches of soil. The next 3 inches contained from 7.0 to 10.5 ppm. After 5 years of spraying, soil from New York peach orchards contained 105 to 116 ppm in the upper 3 inches.

DDT residues in vineyard soils were investigated by Taschenberg, *et al.* (86). A gravelly-loam vineyard had been sprayed with insecticide at the rate of about 6 lb per acre per year for 12 years. The upper 3 inches of soil sampled from along rows

contained 31.4 ppm or 26.9 lb per acre of DDT and 5.4 ppm or 4.4 lb per acre of DDE. The next 3 inches of soil along rows contained 1.5 ppm or 1.5 lb per acre of DDT and 0.05 ppm or 0.05 lb per acre of DDE. Samples of soil taken from the upper 3 inches between rows (where a spray was not applied directly) contained 24.4 ppm or 17.5 lb per acre of DDT and 4.5 ppm or 3.5 lb per acre of DDE. No decomposition products of DDT other than DDE were detected.

Analyses showed soil from fields in which potato crops were grown and sprayed with DDT for an 8 year period contained 12.2 lb insecticide per acre. Similar results were obtained from tests on soils in which treated corn was grown for 3-6 years (32). Ginsburg and Reed (33) found most DDT in orchard soils to be located in the upper 4 inches. Less was found in the next 4 inches and still less in the 8-12 inch depths. Cranberry bogs were found to contain 34.5 lb DDT per acre of which 32 lb was in the upper 4 inches of soil. Corn soils contained an average of 11 lb DDT per acre and potato soils an average of 7 lb per acre. Similar data were also reported by Lichtenstein in 1957 (54). Investigations on other insecticides showed that benzene hexachloride, toxaphene, chlordane, aldrin and dieldrin persisted in sandy loam for at least 3 years although toxaphene and benzene hexachloride appeared to lose a large portion of their toxicity during this period (87).

Lichtenstein applied aldrin and lindane to silt loam and muck soils (56). Seventeen months later 84 to 96% of the insecticides were found in the upper 3-inch layer of loam soil, 4-12% in the next 3 inches and up to 5% in the 6-9-inch layer. Muck soil showed leaching to a somewhat greater degree. Both aldrin and lindane, under non-leaching conditions, moved from treated to untreated soils. Laboratory tests (59) showed lindane was broken down into a non-toxic compound 2 weeks after application to soil. The non-toxic compound was detected by colorimetric lindane tests but not by a bioassay. An average of 26% of heptachlor applied to four different soils (sandy, silt and clay loams, muck) was found present after 21 months (96).

Various chlorinated hydrocarbon insecticides have been used to control Japanese beetle grubs. Tests

<sup>1</sup>Last of a series of three review papers on this subject. For the first and second of the series see, J. Milk and Food Technol., 25: P. 36, P. 72. 1961.

on persistence of these compounds in soil showed an average of 15% of applied chlordane to be present 12 years after treatment (58). Eleven years after treatment, 41% of added benzene hexachloride was recovered by chemical tests but only 8% with a bioassay procedure. No heptachlor was recovered from soils treated with it 9 years earlier when chemical tests were used. A bioassay procedure indicated the presence of 4-5% of the applied toxicant. This was found to be heptachlor epoxide. Most of the aldrin disappeared during a 4-year period. About 8-10% of the applied dosage was recovered as dieldrin.

Factors affecting loss of chlorinated hydrocarbons from soil have been studied by different investigators (61, 63, 71, 72). DDT, endrin and aldrin were applied to soil at the rates of 20, 4.8 and 18 lb per acre respectively (72). Temperatures of the soil surface during the test on aldrin ranged from 52°F at night to 140°F at noon and during the test on DDT and endrin from about 38°F to 125°F. A loss in DDT of 38%, in endrin of 70% and in aldrin of 57% occurred during the first 48 hours after treatment, according to gnat bioassay procedures. A mosquito larvae bioassay procedure showed lower losses of DDT and aldrin but a greater loss of endrin during the same time period.

Laboratory studies by Mulla (71) on stability of insecticides in sandy soil held at 88°F with 11-12% moisture showed no loss in activity of DDT, endrin or lindane after 9 months of storage. No loss of dieldrin was noted after 12 months. After 3 months of storage, 53% and after 9 months, 77% of aldrin had disappeared.

Lichtenstein, *et al.* (63) and Lichtenstein and Schulz (61) studied the persistence of DDT, aldrin and lindane in different soils. General observations made were: (a) DDT was most persistent and aldrin least, (b) insecticides persisted longer in muck soils than in loams, (c) loams of Ohio and Wisconsin retained insecticides to a greater extent than those in Kansas, and (d) insecticides disappeared from soils most rapidly during the first 6 months after treatment. Soil temperature was found to be an important factor affecting persistence of insecticides (61). No loss was found in frozen soils. At a temperature of 6°C, 16 to 27% of aldrin and heptachlor were lost during 56 days. When the temperature was increased to 46°C, losses of 86 to 98% were noted for these two chemicals.

Aldrin and heptachlor in soil have both been found to epoxidize (5, 7, 25). The first is converted to dieldrin and the second to heptachlor epoxide. Such conversions may be of interest since the compounds produced are more persistent than the parent materials. Other studies conducted on these conversions (60) showed when aldrin was applied to quartz sand, Plainfield sand, Carrington

loam and muck soils it was readily transformed into dieldrin in loam but less rapidly in muck soil. The quantities of aldrin and dieldrin recovered from loams were equal 3.3 months after treatment under laboratory conditions (soil held at 37°C) and after 16 months under field conditions. Conversion of aldrin to dieldrin was small in soils with low numbers of microorganisms (Plainfield sand) or with a low moisture content. When heptachlor was applied to loam, it persisted slightly longer than aldrin but the amount of heptachlor-epoxide formed was smaller than that of dieldrin.

Laboratory experiments (59) indicated no dieldrin was formed in aldrin-treated loam when soil was held at 7°C for two weeks. In soils held at 26°C or 46°C, 4-8% of recovered insecticide proved to be dieldrin. The peak of dieldrin formation was reached 56 days after treatment after which a decrease was noted.

Several investigators have shown that insecticides will be deposited in crops grown on contaminated soils (53, 55, 62, 87). Heptachlor was found deposited in potatoes grown in soil the same year it was treated. Aldrin appeared in potatoes for 2 years, chlordane and dieldrin for 3 years after soil was treated (87).

When carrots were grown in lindane treated Miami silt loam, 7.7 times more insecticide was found in the edible part of the carrot (13.9 ppm) than in soil (1.8 ppm). Other crops contained less than soil in which they grew. Tomato fruits contained the smallest amount of lindane. Peas grown in Miami silt loam contained 18 times more lindane in their vines than in their pods. Lindane was absorbed by crops to a greater extent from sandy soils than from soils high in organic matter although the insecticide persisted longer in the latter type.

When different crops were grown in aldrin or heptachlor-treated soils, it was noted that very little, if any, of the two insecticides appeared in onions or seeds of beans or peas. Highest levels (15.9% of aldrin and 31.7% of heptachlor residues in soil) were found in carrots. Carrots were followed in descending order by potatoes, radishes, lettuce, beets, cucumbers and alfalfa (55).

Kenland red clover was grown in clay and silt loams treated with 0.1, 1.0, 10 and 100 ppm of benzene hexachloride (9). Analyses of soils and clovers showed residues in soils, in the sequence listed above, of 0.08, 0.16, 0.91 and 13.84 ppm. Clover grown in the soils contained benzene hexachloride residues of 0.10, 0.24, 2.07 and 14.53 ppm respectively. Dosages up to 200 ppm in soil did not affect germination of clover.

As little as one ppm of crude benzene hexachloride or 8 oz of the gamma isomer per acre in white sand was sufficient to incite root malformation in

Norway pine seedlings (82). The addition of 2.4% of organic matter to white sand offset the phytotoxic effect of benzene hexachloride. Chlordane was non-toxic to conifers at dosages as high as 100 lb per acre in Plainfield sand.

Since soil-borne insecticides have been found to influence growth of plants it was thought they might exert an influence on microorganisms in the soil. Kiigemagi, *et al.* (50) found aldrin and dieldrin contributed little to changes in numbers of soil microorganisms and that soil microorganisms had little effect on the break-down of these two chemicals. Benzene hexachloride, when applied at the rate of one lb gamma isomer per acre, reduced the frequency with which Norway pine seedlings became infected by the damping-off fungus (81). Chlordane had no effect on the fungus. Benzene hexachloride or chlordane had no effect on nodulation of black locust seedlings when chemicals were added to soil at the rates of one and 10 lb per acre, respectively.

The fungitoxicity of lindane (gamma isomer of benzene hexachloride) has been explained by its high solubility in water (77). Chlordane, aldrin and heptachlor are also toxic to fungi and this results from their high vapor point. The low vapor point and the relative insolubility in water of methoxychlor, dieldrin and endrin make them relatively non-toxic to fungi.

Eno (19) reviewed effects of different chlorinated hydrocarbons on soil microorganisms. His discussion of DDT indicated: (a) CO<sub>2</sub> evolution and dextrose decomposition in soil were not reduced by addition of 137.5 lb DDT per acre, (b) applications of 20 lb DDT per acre had no harmful effect on soil microorganisms in various loams, (c) 103 lb DDT per acre increased rod length and frequency with which gram positive cells of *Rhizobium leguminosarum* appeared in root nodules of the common bean, (d) number of nodules per plant (common bean) was depressed by 103 lb DDT per acre, (e) 0.01 to 0.001% DDT was not toxic to nitrifiers, ammonifiers or sulfur oxidizing microorganisms although high insecticide concentrations caused injury to nitrifiers and ammonifiers, and (f) DDT in sandy soil (up to 120 ppm) resulted in significant increases in numbers of bacteria, actinomycetes and fungi but increases were not proportional to amount of DDT added.

A summary of information about benzene hexachloride indicated: (a) the gamma isomer increased numbers of bacteria but inhibited *Streptomyces*, (b) delta and gamma isomers increased molds in dextrose enriched soils while alpha and beta forms depressed them, (c) delta and gamma isomers increased ammonification of peptone; beta and gamma isomers increased nitrification, (d) numbers of *Azotobacter* were reduced by 4 lb of 20% benzene hexachloride per acre and (e) nodulation on red clover, soybeans, al-

falfa and hairy vetch was seriously inhibited by 30 ppm of benzene hexachloride. Chlordane had no significant effect on ammonification and nitrification when 200 ppm were in soil but it did increase numbers of bacteria and reduce numbers of fungi when 50 ppm were present. Concentrations of aldrin as low as 25 ppm depressed nitrogen transformations in soil. In several soils 200 and 1,000 ppm had a stimulatory effect on total numbers of microorganisms. No appreciable effects were noted on microorganisms when toxaphene, dieldrin or heptachlor were added to soil.

Alexander (1), in his recent book on soil microbiology, summarizes the effect of insecticides on soil microorganisms when he says: "Little or no inhibition (of soil microorganisms) is found as a result of using DDT, benzene hexachloride, chlordane, aldrin, parathion and toxaphene. Among the more sensitive processes to such compounds are nitrification and legume nodulation; where damage is noted, it is usually upon one or both of these two. However, it is unlikely that the inhibitions would materially affect crop production, particularly when weighed against benefits accruing from proper usage of the pesticides."

Lakes may become contaminated with chlorinated hydrocarbons through surface run-off from treated soils or through direct application to control a particular pest. A fresh water lake in California was treated with DDD in 1949, 1954 and 1957 to control gnats (44). Studies on specimens from the lake were begun in March of 1958 and showed: (a) all fish, bird and frog samples analyzed contained DDD, (b) the amount of DDD found in flesh samples was greater than that in lake water on a ppm basis, (c) flesh samples of largemouth bass and Sacramento blackfish hatched 7 to 9 months after the last DDD application contained 22 to 25 ppm and 7-9 ppm DDD, respectively and (d) all areas of the lake contained fish contaminated with DDD. Analysis of visceral fat from apparently healthy largemouth bass and white catfish indicated accumulations of DDD at levels as high as 2,275 ppm and 1,700 ppm respectively. Grebes apparently could not tolerate the same high concentration of insecticide since many died of chronic DDD poisoning.

#### SUMMARY

Chlorinated hydrocarbons may be deposited in a variety of plant tissue after spraying or dusting treatments. Additional deposition may result if plants grow in previously contaminated soils. Vegetables, fruits, nuts, wheat and tobacco are crops used for human consumption which have been found to contain chlorinated hydrocarbon residues. Pastures, hay crops, and silages may contain insecticide residues

which are ingested by cattle when the crop is consumed. Many plants appear able to convert aldrin into dieldrin and heptachlor into heptachlor epoxide.

Livestock may be exposed to chlorinated hydrocarbons through spray and dust treatments or through ingestion of contaminated feed. The insecticides appear in both milk and meat of dairy cattle. Residues in meat are generally greater in these cuts with a high proportion of fat. High-fat dairy products made from contaminated milk contain substantial quantities of insecticides. The tissues (primarily fatty) of swine, sheep and poultry have also been found to contain various levels of insecticides after animals had been previously exposed to the compounds.

Chlorinated hydrocarbons have been deposited in eggs by hens after ingesting insecticides. Highest residue levels occurred in the yolk portion of eggs. Soils may become contaminated with insecticides either by chance when plants are treated or directly when the compound is added to control some soil-borne pest. Chlorinated hydrocarbons in soil persist for many years and, during that time, may be absorbed, in part, by various crops. Aldrin and heptachlor appear to epoxidize in soil and form dieldrin and heptachlor epoxide, respectively.

A study on use of DDD to control gnats around a fresh water lake indicated the compound was absorbed and retained by tissues of fish and frogs. Fatty tissue from certain fish contained unusually high levels of insecticide while chronic poisoning of others was observed.

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