PESTICIDE RESIDUES IN SOILS'

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The pesticide situation

The forest nurseryman has greatly increased his use of pesticides in recent years through necessity, just as have all other persons and agencies whose livelihood or duties require the controlling of pests. The term pesticide broadly includes compounds intended for a variety of purposes. They are used to control insects, diseases, weeds, rodents, pest birds, predatory animals, and rough fish. Plant growth regulators, defoliants, and desiccants are also included under the term pesticides. If the term agricultural chemicals is used, this would also include chemical fertilizers.

In 1961, over 500 million pounds of pesticides worth 300 million dollars were sold by United States companies. There are approximately 94,000 brand-name registered pesticides on the market. These chemicals are distributed annually over nearly 200 million acres (but this is still only about 1 acre in 12 treated within the United States). Along with mechanization, increased use of chemical power to kill pests and do work has enabled the American farmer to give us the most healthful and plentiful food supply of any nation in the world. This food is available to us at a less cost in terms of work hours than ever before. For example, the American family spends 20 percent of its income on food. By contrast, the Russian family spends over 50 percent; in Italy and Peru nearly 40 percent; and in Nigeria, 70 percent. Forest insects and diseases cause greater timber losses than by forest fires and n11 other agencies combined. They destroy an estimated 7 billion board feet of timber a year. Foresters predict that the nation's 489 million acres of commercial forest will someday be as intensively cultivated as our nearly 400 million acres of cropland are today.

Chemicals as pesticides are here to stay. An increasing population will require more plants to be grown for food, fiber, and shelter from a shrinking agricultural acreage. Without pesticides, according to a National Academy of Science Sub-Committee, many fruits and vegetables would disappear from the market, and many other crops would grow so meagerly they would be marketable only as rare luxuries. In addition, pesticides allow profitable yields of nonfood crops, such as cotton,

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tobacco, and timber. The President's Science Advisory Committee in its report on the use of pesticides recognized the necessity of pesticides, but also suggested that there are apparent risks.

One of the problems which needs further study, research, and clarification is the persistence of pesticides in the environment, including persistence in soils. For instance, DDT has been detected at great distances from the place of application and its concentration in certain living organisms, including man, has been observed. Small amounts of DDT have been detected in food from many parts of the world. The amounts are rarely above Food and Drug Administration (FDA) tolerance limits, but these have probably contributed to the buildup of DDT we now observe in the fat of the people of the United States, Canada, Germany, and England.

Responsibilities of the forest nurserymen

Thus, even though the forest nurserymen are not primarily concerned with pesticide applications to food or feed crops, they should be aware of the necessity, value, and proper use of pesticides. The applicator's safety needs to be safeguarded, injury to seedling trees prevented, and buildup of harmful residues avoided. Those nurseries growing food or feed crops definitely need to be aware of laws concerning use of pesticides on these crops and of proper usage of the chemicals.

Laws regulating agricultural chemicals

The forest nurseryman, just as any person using or supervising use of pesticides, should be aware of the Federal and State Laws designed to assure the safety of the public.

The basic federal laws regulating pesticides are:

1. The Federal Insecticide, Fungicide, and Rodenticide Act (1947). (A revision of the Insecticide Act of 1910).

2. The Miller Pesticide Residue Amendment (1954) to the Federal Food, Drug, and Cosmetic Act (1938).

The Federal Insecticide, Fungicide, and Rodenticide Act, in general, requires that economic poisons (pesticides) shipped in interstate commerce be registered with the United States Department of Agriculture. The registrant must present data to prove its usefulness, safety to the public when label precautions are followed, compatibility of various crops, and safety of the treated crop product for use as a livestock feed or food for human consumption.

The pesticide manufacturer must furnish pertinent information on composition of the material, rate and timing of application, and adequate precautionary information to protect the user and the general public. If residues on food crops are present, registration is delayed until tolerances are established.

In 1')5y, an amendment (Public Law 86-319) to the Federal Insecticide, Fungicide, and Rodenticide Act expanded the coverage of the Act to include such other agricultural chemicals as nematocides, defoliants, desiccants, and plant growth regulators. The Act was amended again in 1962. Practically **all** chemicals and devises intended for the destruction or repelling of pests were brought under the surveillance of the Department of Agriculture. This amendment declares to be pests, when they are injurious to .marts interests, mammals, birds, fish, amphibians, reptiles, aquatic and terrestrial invertebrates, unwanted roots or other plant parts, and viruses other than those on or in living man or animals.

The Miller Pesticide Residue Amendment (Public Law 83-518) was enacted to protect the public from any harmful residues of pesticide chemicals used in the production, storage, or transportation of raw agricultural commodities.

Under this law a pesticide manufacturer usually files a petition with the Federal Food and Drug Administration requesting establishment of a tolerance or exemption of the particular pesticide residue in a crop or food. The petition must include data on the safety of the chemical, results of toxicity and residue studies, and the proposed tolerance. The Department of Agriculture must certify to the FDA that the chemical has a useful place in agriculture and that the tolerance proposed reasonably reflects the residues likely to result when the pesticide is used as directed.

If FDA is satisfied on the basis of available data that residues within the proposed tolerance are safe, it publishes an order establishing the tolerance. If a product is evaluated and recognized as safe, a tolerance or exemption is not required. irlhen evidence is available that the proposed use of a chemical will leave no residue, the Department of Agriculture registers such products on a "no residue" basis and no tolerance is needed.

Once a tolerance is established, the FDA enforces it. Any raw agricultural commodity found in interstate commerce, or at any time after it has been moved in interstate commerce, with a residue exceeding the legal tolerances, is subject to by FDA agents. Tolerance can also be enforced by criminal proceedings.

The Food Additives Amendment to the Federal Food, Drug, and Cosmetic Act as written in 1958 had no direct effect on growers who use pesticides in the production of their crops. Only those pesticides, such as preservatives and fumigants, which are added to food during or after processing are directly subject to it. This amendment requires that the use of any substance which can reasonably be expected to become a component part of any food, or in any way change the characteristic of any food, must be covered by Food and Drug regulation prescribing how the additives can be safely used. This amendment contains the Delaney "cancer'? clause. The Delaney clause states that no permission will be granted to use an additive, if it has been found to cause cancer in man or animals when fed at any level of intake.

In 1959, by administrative interpretation, this provision was applied to pesticides under the Miller Amendment.

State laws and regulations in most states either duplicate the same federal requirements within the state, or establish similar requirements to protect the safety of users and consumers.

Pesticides and the soil

Many pesticides are applied directly to forest nursery soils to control weeds, plant diseases, insects, and nematodes. Certain pesticides are used routinely in the production of seedlings and some are used only occasionally. Parts of pesticides that are applied to plant foliage also reach the soil. Therefore, knowledge of the behaviour and persistence in soils of these chemicals is essential for continued safe tree seedling production and in some cases production of food crops.

Soil persistence problems and hazards related to pesticide usage in agricultural crop lands may be placed into four categories: (1) injury to crops which follow a sprayed crop in a rotation system, (2) unlawful residues in crops which follow a sprayed crop in a rotation system, (3) accumulation of pesticides from application rates which exceed rates of dissipation from soils, and (4) toxic residues that may injure beneficial soil micro--organisms. With some exception, the general principles of these problems should also apply to forest nursery soils.

Processes involved in pesticide dissipation from soils and possible persistence hazards will be discussed in general terms in this paper. More emphasis will be placed on persistence of herbicides in soil since these chemicals are within the author's area of educational responsibilities. Where possible, persistence of those pesticides applied to forest nursery soils will be discussed.

Factors affecti persistence of pesticides in soil

Pesticides may be dissipated from the soil through the influence of one or several processes. Factors affecting persistence are (1) microbial decomposition, (2) chemical decomposition, (3) adsorption on soil colloids, (4) leaching, (5) volatility, and (6) photo-decomposition. In general, organic pesticides are dissipated more rapidly than inorganics, but some exceptions probably exist.

Inorganics such as arsenates, borates, and chlorates are lost from the soil or become unavailable for uptake by plants, through the formation of insoluble compounds and complexes, leaching, dilution in soil, and fixation and adsorption to soil. Small amounts may be absorbed by plants and lost by removal of plant material from the soil.

Table 1 gives an approximate length of persistence for various herbicides under a given set of conditions. The information is based on experimental work and general observations. In general, the conditions stated are favorable to rapid herbicide decomposition - or a short period of herbicide toxicity.

Pathways of pesticide breakdown

The importance of a process in causing loss of a pesticide from soil will depend on the physical and chemical properties of the chemical, soil properties, and prevailing environmental conditions. Inactivation or decomposition of a pesticide which is active for only a few days to a few weeks is largely due to a single process. Adsorption of some pesticides, for example, is rapid and sufficiently complete to render them immediately inactive when applied to soil. Some nematocides and herbicides disappear from soil within 1 to 3 weeks by volatilization or by reactions and volatilization.

Degradation by soil micro-organisms and leaching from soils may account for loss of a major part of some pesticides that disappear rapidly. The longer a pesticide persists, the greater the probability that several processes become involved in its inactivation and disappearance. This concept is summarized by several examples in table 2.

Decomposition by soil micro-organisms is also an important pathway of loss of many organic pesticides which persist for a few weeks or more in soils. Adaptation of effective soil micro-organisms occurs when chlorinated aliphatic acids such as dalapon, chlorinated phenoxyacetic acids such as 2,4-D, and probably several other groups of pesticidal chemicals are added to soils. Decomposition curves which illustrate that an adaptation is necessary show (a) a lag phase with no appreciable change in concentration, followed by (b) a period of rapid loss. The time required for adaptation varies with the pesticide, species of micro-organisms, and environmental conditions.

The organic pesticides which persist for several months, a long lag period may be necessary for adaptation of effective soil micro-organisms. The long lag period required for 2,4,5-T might explain delayed decomposition of this herbicide under some conditions. However, extended lag periods do not explain the persistence of several s-triazine and phenylurea herbicides, which often persist for several months in soils. These herbicides appear to be decomposed by soil micro-organisms which require no adaptation.

Table 1.--Persistence of herbicides in a moist loam field soil, with little or no leaching, under summertime temperatures in a temperate climate

Chemical	Rate per acre	Type of treatment	Persistence in time
2,4,-D	1/2 - 3 lbs.	pre- and postemergence	1 - 4 weeks
Sesone	2 - 4 lbs.	pre-emergence	2 - 4 weeks
MCPA	1/2 - 3 lbs.	pre- and postemergence	1 - 4 weeks
2,4,5-T	1/2 - 3 lbs.	pre- and postemergence	2 - 5 weeks
Silvex	1/2 - 3 lbs.	pre- and postemergence	2 - 5 weeks
NPA	2 - 8 lbs.	pre-emergence	1 - 4 weeks
2,3,6-TBA	1 - 3 lbs.	pre- and postemergence	2 - 10 weeks
TCA	40 -100 lbs.	postemergence grass control	50 - 90 days
Dalapon	5 - 40 lbs.	postemergence grass control	10 - 60 days
PCP	5 - 20 lbs.	pre- and postemergence	1 - 5 weeks
DNBP	6 - 9 lbs.	pre-emergence	3 - 5 weeks
Amitrol	2 - 10 lbs.	pre- and postemergence	3 - 5 weeks
MH	3 - 6 lbs.	postemergence	1 - 5 weeks
Simazine	1 - 4 lbs.	pre-emergence	3 - 6 months
Simazine	10 - 40 lbs.	soil sterilant	6 - 24 months
Fenuron	4 - 40 lbs.	soil sterilant	3 - 12 months
Monuron	1 - 3 lbs.	pre-emergence	3 - 6 months
Monuron	20 - 50 lbs.	soil sterilant	6 - 20 months
Diuron	1 - 3 lbs.	pre-emergence	3 - 6 months
Diuron	10 - 40 lbs.	soil sterilant	6 - 24 months
Neburon	2 - 8 lbs.	pre-emergence	3 - 6 months
IPC	4 - 8 lbs.	pre-emergence	2 - 4 weeks
CIPC	4 - 8 lbs.	pre-emergence	3 - 5 weeks
CDEC	4 - 8 lbs.	pre-emergence	3 - 5 weeks
EPTC	2 - 6 lbs.	pre-emergence	3 - 8 weeks
CDAA	4 - 8 lbs.	pre-emergence	3 - 5 weeks
Calcium cyanamide	400-4000 lbs.	pre-emergence	1 - 5 weeks
Potassium cyanate	8 - 16 lbs.	postemergence	none
Ammonium sulfamate	100-400 lbs.	postemergence	1 - 3 weeks
Sodium chlorate	450-1200 lbs.	soil sterilant	6 - 12 months
Arsenic	750-1000 lbs. (As_2O_3 equiv.)	soil sterilant	6 - 24 months
Boron	1000-3000 lbs. (B_2O_3 equiv.)	soil sterilant	6 - 24 months
Hydrocarbon oils	50 -400 gals.	postemergence	usually none; however, varies with oils

(Taken from Klingman, Glenn C., Weed Control: As a Science. John Wiley & Sons, Inc., New York. 1961)

Table 2.--Relation of disappearance time of pesticides to the number of processes which cause inactivation and decomposition

Pesticide	Disappearance of inactivation time (weeks)	Dissipation on processes in decreasing order of relative importance
Diquat	0	adsorption
Methyl bromide	0 - 0.3	volatilization
SMDC	1 - 3	chemical reaction, volatilization
Dalapon, 2,4,-D 2-chloro-4-(hydroxy-mercuri) phenol	1 - 3	microbial decomposition, dilution, and leaching
EPTC	3 -10	volatilization, microbial metabolism, and adsorption
Diuron	6 -24	adsorption, microbial decomposition, dilution, leaching, photodecomposition, and uptake by higher plants
Atrazine	6 -24	adsorption, microbial decomposition, volatilization, dilution, leaching, and uptake by higher plants
DDT, BHC, aldrin, dieldrin, toxaphene	8 -52	adsorption, volatilization, chemical reactions, dilution, leaching, uptake by higher plants, and erosion
Arsenates, borates, chlorates	26 -52	chemical reactions that give insoluble compounds and complexes, dilution, leaching, and adsorption

(Taken from Sheets, T. J., Dissipation of Pesticides From Soils. Proceedings North Carolina Soil Science Society, January, 1964).

Disappearance curves for phenylurea and s-triazine herbicides show that decomposition begins soon after contact with the soil, but the rate of disappearance gradually decreases with time. The rate of decomposition appears to be a function of the concentration (rate of application) in the soil. The greater the rate of application, the greater the amount of herbicide decomposed. The amount decomposed per unit applied remains unchanged.

Possible persistence hazards

Rates of disappearance of several chlorinated hydrocarbons from soils vary. In several midwestern soils, studies showed that DDT was more persistent than aldrin. Lindane disappeared most rapidly. Dieldrin was recovered as a persistent residue from several aldrin-treated soils. In studies on the accumulation of nine insecticides in field soils at Beltsville, Maryland; State College, Mississippi; and New Brunswick, New Jersey, the tendency for accumulation was greatest at Beltsville and least at State College. Differences in persistence were probably due to variations in soil properties and environmental conditions.

Some researchers report little or no accumulation in soil of chlorinated hydrocarbons applied at rates used for insect control. In one study, one week after the sixth weekly application of heptachlor at 0.33 pounds per acre, only 31 percent of the total application remained. Other data, however, indicate that several chlorinated hydrocarbon insecticides (DDT, endrin, toxaphene, and possibly dieldrin) are rather persistent and may accumulate to at least some extent under some conditions from repeated applications. In southern California, 22 to 25 ppm of DDT or toxaphene accumulated in soils sprayed annually for 5 years with 20 pounds per acre. Residues decreased at about 25 percent per year after the applications stopped. Dieldrin, endrin, chlordane, and heptachlor at 5 to 10 pounds per acre per year accumulated to 3 to 5 ppm and disappeared at about the same rate as DDT and toxaphene after applications were stopped. Aldrin and lindane did not accumulate.

From data available, generalizations on the magnitude of the hazards of accumulation of the chlorinated hydrocarbons are difficult. High levels of BHC, DDT, aldrin, and chlordane residues have decreased yields and affected quality of crop plants. Forest nurserymen using appreciable quantities or repeated applications of these pesticides should seek further information on possible residues in soils.

Most organic phosphates, although very toxic to insects and to many warm-blooded animals, are not toxic to soil micro-organisms and are rapidly dissipated from soils. At rates used for pest control, carbonates, thiocarbamates, and thiolcarbamates sometimes persist for several weeks in soil, but as a group they have not caused significant persistence problems.

The chemicals most widely used as nematocides are ethylene dibromide (EDB), 1-2 dibro-3-chloropropane (DBCP), and 1,3-dichloropropene, obtainable either as technical dichloropropenes or mixed with 1,2-dichloropropane (D-D mixture). All these nematocides are more or less toxic to plants when applied to the soil, but after several days or two weeks this effect is no longer noticeable for most plants. Length of persistence depends on the chemical, the crop plant, and the soil type.

Ethylene dibromide leaves in the soil a residue containing bromine which is taken up by various plants. A number of tolerances for bromine in vegetables and fruits grown in soil fumigated with EDB have been set by FDA.

Dichioropropenes and D-D mixtures hydrolyze and form hydrochloric acid in the soil, but other breakdown products or residues in soil or plants under normal conditions of use have not been reported. However, it appears that plants can take up something derived from these materials since toxicity symptoms appear if various kinds of plants are planted too soon after application.

Soil fumigated with methyl bromide has a residue containing bromine. This is sometimes troublesome with plants easily injured by bromine. However, most crop plants can be planted without injury in methyl bromide-treated soil 72 hours after the cover has been removed. Methyl bromide is registered with USDA on a nonresidue basis for preplant soil treatment at 2 pounds per 100 square feet with an aeration period of 72 hours.

Since methyl bromide and certain other soil fumigants may kill beneficial soil organisms as well as soil pests, indirect undesirable consequences may result from soil fumigation with these materials. Normal decomposition of organic matter into ammonia, then nitrite, and finally nitrate may be disturbed so that toxic substances accumulate.

For example, in sterilized soils high in organic matter, ammonia or nitrite may build up to toxic levels for plants. Usually the beneficial micro-organisms multiply sufficiently in 1 to 3 months time in warm, moist soil so as they restore a normal micro-organism equilibrium and normal plant growth. Forest nurserymen are probably aware of these problems since they routinely use methyl bromide.

Although some benzoic acid herbicides such as amiben disappear from soils within 2 to 3 months, 2,3-6-TDA persists longer. At rates used for witchweed control in corn, 2,3-6-TBA and fenac were active [illegible]

Fenac has injured tobacco 4 years after initial application to the soil.

Herbicides in the phenylurea family (diuron, monuron, etc.) and s-triazine family (Simazine, atrazine, etc.) can persist from one season to the next and cause injury to sensitive plants. However, when label instructions are followed and the materials are properly applied within recommended rates, there has been no widespread carry-over problem in North Carolina. Misuse of the chemicals has largely been the cause of residue problems where they have occurred. Sometimes diuron applied to cotton may persist and injure cereals planted in the fall after harvest. Lay-by applications of diuron or pre-emergence applications plus lay-by applications prevent fall-seeded crops from being planted in treated fields and sensitive crops can not be planted the following spring.

Atrazine and simazine applied pre-emergence for selective weed control in corn sometimes persist and injure sensitive crops the next year. This has been more troublesome in the northern plain states and Canada than in the southeast.

Diphenamid appears to have the potential for carry-over into the fall at the higher rates on some soil types. Cereal crops would be sensitive to these soil residues. Trifluralin, although a long-lasting herbicide, has not given soil residue problems in use to date.

The hydrocarbon oils (Stoddard solvents and varsol) commonly used by forest nurserymen present no residue hazards. These oils evaporate quickly. Residues of petroleum solvents in most crops are exempt from requiring tolerances.

Inactivation rate of several herbicides including diuron and simazine are related to the initial concentrations of the chemical in the soils. Theroretical calculations based on this interpretation show possible accumulation levels in soils. If 80 percent (this loss is a reasonable minimum rate of loss for these herbicides) of an annual application of 2.0 pounds per acre is inactivated each year and this application is repeated indefinitely on the same soil, the amount present in the soil prior to each application would eventually approach 0.5 pounds per acre. If the annual decomposition is 50 percent of the total at the beginning of the year, the amount in the soil just before the annual application would eventually approach the amount added each year. Rates of inactivation of herbicides vary tremendously with chemical structures, soils, and climates. Therefore, carry-over of the persistent selective herbicides can be expected under some conditions but the possibilities of hazardous accumulations from proper applications do not seem likely.

Measures to *minimize* or avoid residue problems

These measures are given in terms of herbicides on cropland and in cropping sequences but many of the principles will apply to forest nursery soil pesticide use.

1. When a crop is grown continuously, such as corn following corn treated with atrazine, rotate herbicides on the same crop. For example, if atrazine is used as a pre-emergence herbicide on corn, a less residual type herbicide would be used the year preceding a different crop.

2. Rotate the use of herbicides when the same crop is grown continuously but also rotate the use of herbicides on all crops grown in the rotation. For example, if cotton and corn are grown in rotation, use atrazine one year on corn and a less residual type herbicide on corn the next time it appears in the rotation. Also, if diuron is used on cotton the first time it appears in the rotation, use a less residual type herbicide the next time it is planted in the rotation. The practice aids in controlling a broader spectrum of weeds and reduces the likelihood of residue problems.

3. Select a cropping sequence which employs crops that are tolerant to the herbicide. For example, a spring sown grain crop may be planted rather than a fall sown crop. In other instances the tolerant crop, corn treated with atrazine for example, can be grown continuously until the weed population is reduced.

4. Use the lowest rate of each herbicide to obtain the degree of weed control required. Supplement this with effective cultural practices. Band treatment instead of broadcast application will add less total chemical to the soil.

5. Use mixtures of herbicides that contain the minimum amount of each herbicide to control those weeds on which each herbicide in the mixture is most effective. For example, a mixture of 4,6-dinitro-0-sec-butylphenol (DNBP) at 1 pound per acre and sodium 2,4-dichlorophenoxyethyl sulfate (sesone) at 3 pounds per acre is more effective for weed control in peanuts than ti pounds per acre of DNBP or 6 pounds per acre of sesone used alone. A mixture of diphenamid and trifluralin also exhibits the same properties.

6. Liquid formulations of some soil-applied residual type herbicides are often less residual than granular formulations of the same herbicides, depending on the materials used in their formulation.

7. Liquid formulations of residual type soil-applied herbicides can be applied more uniformly with present equipment available than granular formulations with present granular application equipment.

8. Herbicide residues from applications made at planting time are less likely to cause damage than residues from an equal amount of herbicide applied later in the season as a postemergence spray.

V. Deep plowing followed by thorough disking and thorough seedbed preparation before planting the succeeding crop after a herbicide has been used on preceding crops will reduce the risk of injury from herbicide residues.

10. Uniform application of the herbicide is extremely important. The spray pattern from sprayer or applicator should not overlap. Spray equipment should be operated at constant speeds and double coverage should be avoided. Use low pressures to lessen danger of spray drift.

11. The sprayer should be equipped with check valve nozzles and a quick closing valve to prevent drip. The sprayer should be cut off on head rows and turn rows and in other areas where overdosage is likely to occur. Use proper nozzles and correct screen size for nozzle and material being applied.

12. READ AND HEED THE LABEL. If the forest nurseryman follows labels in using **all** pesticides, there appears to be very little possibility that his soils will accumulate residues that will cause any problems.

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