Chapter 10

EFFECT OF SOIL PROPERTIES AND PROCESSES ON HERBICIDE PERFORMANCE AND PERSISTENCE

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INTRODUCTION

Soil-applied herbicides are designed to achieve continuous weed control over periods of time ranging from a few weeks to several months. Effective herbicides should persist in soil at concentrations that will provide weed control but should not persist long enough for carryover to injure following crops. Increased persistence also increases the opportunity for movement of herbicide into surface or groundwater. Herbicide performance and persistence are determined by the properties of the herbicide, the soil and climatic factors and the physical, chemical and biological processes to which these chemicals are subject. Understanding these factors and processes can result in improved and more effective methods of herbicide use, and aid in the development of new herbicides while minimizing undesirable contamination of soils and waters.

Knowledge of herbicide-soil interactions is needed at several different levels. Cotton farmers seek to match weed control needs with effective chemicals on a field-specific basis. Equally specific knowledge of the soils and a reasonable anticipation of weather are taken into account in the selection of herbicides. Weed scientists seek to integrate knowledge of herbicide chemistry, soil interactions and environmental effects to develop new application techniques and to identify herbicides that will provide effective weed control over broad regions. Scientists concerned with meeting or developing regulations that protect the environment require both broad knowledge of these interactions and specialized knowledge, including herbicide persistence and movement at trace concentrations, mechanisms of degradation and the identity and toxicological properties of herbicide metabolites.

ENVIRONMENTAL FACTORS AFFECTING HERBICIDES IN COTTON SOILS

The environmental factors of climate and soil primarily affect the retention, transformation and transport processes that control herbicide behavior in soil. The climatic factors include temperature, moisture, air movement, humidity and solar radiation; soil factors include organic matter and clay content, pH and salinity. Due to the interactions between climatic and soil factors and the processes affecting herbicides, it is often difficult to isolate the effect of any one factor on a given process. Therefore, only generalities concerning environmental factors based on a number of review articles will be discussed here (Guenzi, 1974; Hance, 1980b). More specific details will be discussed later in this chapter.

CLIMATE

Cotton is grown in the United States south of 37° latitude due to a direct relationship between potential yield and growing days. The crop requires at least 160 days above 60F in order to obtain maximum yields. A second limiting factor for cotton yield is the total amount and distribution of rainfall and irrigation during the growing season. Cotton is grown in areas of the United States with rainfall ranging from 6 inches in the West to 60 inches in the Southeast (Waddle, 1984). The cotton grown in low rainfall (6-inch) areas is irrigated. These variations in temperature and moisture regimes also affect performance and persistence of herbicides over the Cotton Belt.

The herbicide properties of solubility and vapor pressure and the processes of degradation and volatilization are dependent on temperature with both properties and processes increasing as temperature increases. High temperatures can lead to increased herbicide losses through increased degradation and volatilization. Low humidity and high temperatures dry out soils which may decrease plant uptake of herbicides by reducing movement of herbicide to plant roots and increasing loss of herbicide by increasing volatility.

The amount, intensity and frequency of rainfall or irrigation water controls the movement of pesticides to and away from target plants and the ability of the pesticide to go into solution. Under dry conditions, some precipitation is necessary to move soil-applied herbicides to the rooting zone where uptake can occur. Heavy rains immediately after herbicide application can lead to surface runoff of the herbicide. A large quantity of rainfall at low intensities can move some herbicides past the rooting zone and decrease herbicide effectiveness.

Degradation is also primarily dependent on temperature and moisture conditions, thereby affecting the persistence of a herbicide. Degradation tends to increase under warm, moist conditions while decreasing under dry and cold conditions. For instance, Burnside *et al.* (1963, 1969) showed that triazine, *i.e.* atrazine (AAtrex®), and substituted-urea, *i.e.* monuron (Telvar®), herbicides' persistence in the wetter eastern part of Nebraska was less than in the drier

western part. Various studies have shown that herbicides are more persistent in the cooler northern and drier western states than in the warmer wetter southeastern states. Under normal conditions there is sufficient oxygen in the soil for aerobic degradation to take place. However, under extremely wet conditions where there is no oxygen, anaerobic degradation may replace aerobic degradation, depending on the presence and ability of anaerobic organisms to degrade a herbicide.

SOIL

Cotton can be grown on any soil used to grow crops in the United States below 37° N latitude that is subject to the aforementioned temperature and moisture regimes. Cotton is grown on seven major soil orders and 20 subgroups. These soils have as wide a range in properties—organic matter contents, pH levels, salinity levels and clay contents—as do soils used for other row crops. In general, the soils used for cotton production have a lower organic matter content than soils with other crops primarily due to the environmental conditions of the cotton growing areas. High soil temperatures and moisture contents, combined with intensive tillage, accelerate organic matter decomposition.

There have been numerous attempts to correlate herbicide performance and persistence to soil type (*i.e.* as a function of soil texture, organic matter content, pH). However, the reports in the literature are conflicting. Herbicide persistence, for instance, has been reported to increase, decrease or remain the same in soils with increasing soil organic matter content (Hurle and Walker, 1980). The reasons for the conflicting reports could be the differences in physical and chemical properties among the soils, differences in herbicide chemistries used in the studies, and differences in mechanisms of herbicide degradation.

Soil organic matter and clay content are probably the most important soil factors that affect herbicide performance and persistence. They indirectly influence all the processes affecting herbicides. As a rule, the greater the organic matter and clay contents, the greater the adsorption of the herbicide to the soil particles. Adsorption removes the herbicide from solution or air, thereby making less chemical available for movement in water and air, decomposition by microorganisms and absorption by plants. In essence, it lowers the amount of herbicide that is actively available and hence lowers its performance. Crop residues on the soil surface in reduced tillage systems can also adsorb herbicides—as much as 50 percent of the total amount applied in some cases (Banks and Robinson, 1984). Also, in soils with higher organic matter contents, microbial populations are usually greater which in turn may lead to greater rates of degradation of the herbicide.

Soil pH can affect both the physical and chemical properties of some herbicides and some processes. Increased solubility occurs at solution pH values where *s*-triazine herbicides are protonated (positively charged cation) or weak acid herbicides are dissociated (negatively charged anion). Soil pH also affects

the decomposition and movement of different herbicides. For example, hydrolysis (one form of chemical decomposition) of chlorsulfuron (Glean®) is more rapid in acidic soils (pH < 7) (Joshi *et al.*, 1985). Acidic herbicides are more mobile at pH values where the molecule exists as the anion.

SOIL PROCESSES AFFECTING HERBICIDE ACTIVITY IN SOIL

For successful weed control using soil-applied herbicides, levels of herbicide must remain available in the soil long enough for the seedling to absorb toxic levels of the chemical. Once applied to soil, three processes reduce the availability of the herbicide to the target plant: retention, transformation and transport. These processes are affected by the pesticide properties, by the soil and by environmental factors (Figure 1) acting in concert to control the amount of available chemical in solution at a given time.
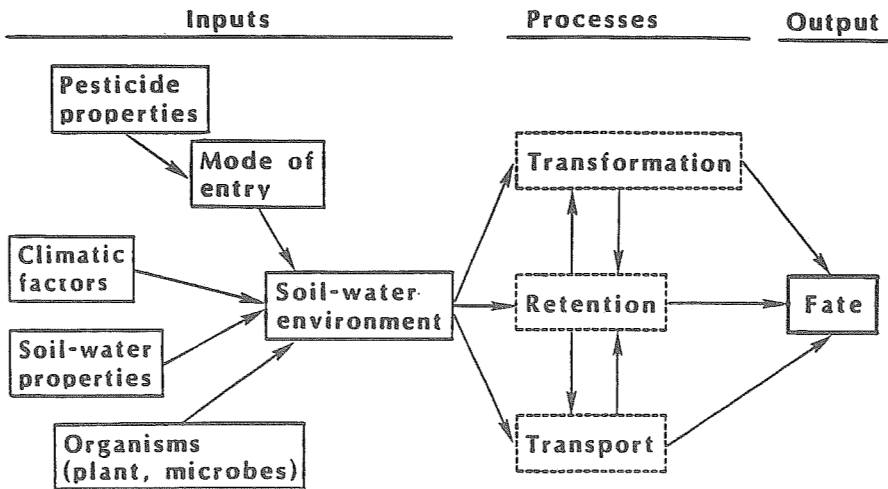

Figure 1. Interaction of the factors and processes affecting the rate of pesticides in the soil environment. (From Cheng and Koskinen, 1986.)

HERBICIDE RETENTION PROCESSES

Retention is a primary factor affecting the efficacy of soil-applied herbicides. The retention process refers to the ability of the soil to hold a herbicide and prevent the chemical from moving. Retention also controls the amount of herbicide available for transformation processes. Retention primarily refers to the adsorption process, but also includes the absorption of herbicides by the soil

particles, plants, and microorganisms. The literature abounds with references on the retention of pesticides in soils (*e.g.*, Hamaker and Thompson, 1972; Green, 1974; Weed and Weber, 1974; Calvet, 1980; and references cited therein). This section will present some of the main features and generalizations concerning the adsorption-desorption process.

Herbicides can be adsorbed to soil by a number of mechanisms including physical bonding such as van der Waals forces, hydrogen bonding and dipole-dipole interactions, chemical bonding such as ion exchange, covalent bonding, protonation, ligand exchange, cation bridging and water bridging with varying degrees of strengths of interactions. For hydrophobic, nonpolar compounds with low water solubilities, adsorption also has been described as a hydrophobic partitioning process between soil water and a soil organic matter phase (Table 1).

Adsorption of nonionic, nonpolar hydrophobic herbicides is due to weak attractive interaction such as van der Waals forces. Electrostatic interactions can also be important, especially when a herbicide molecule is polar in nature. Attraction potential can develop between polar molecules and the heterogeneous soil surface that has ionic and polar sites, resulting in stronger bonding. For any given herbicide or family of chemicals, there is likely a continuum of mechanisms responsible for adsorption onto soil particles. For instance, within the triazine herbicide family, it has been suggested that mechanisms involving van der Waals forces, charge transfer, hydrophobic bonds, cation exchange and cation bridging are responsible for bonding to soil surfaces (Hayes, 1970).

Adsorption of phenyl- and other substituted-urea herbicides to soil or soil components also has been attributed to a variety of mechanisms depending on the adsorbent. The mechanisms have included hydrophobic bonding (Carringer *et al.*, 1975), cation bridging (Hance, 1971; Gaillardon *et al.*, 1980), van der Waals forces (Hance, 1969; Senesi and Testini, 1980) and charge-transfer (Hance, 1969; Senesi and Testini, 1980). Even cation exchange has been reported for the substituted-urea tebuthiuron (Spike®) (Weber, 1980).

The degree of herbicide adsorption is controlled by a number of factors including amounts and types of soil organic matter and clays, herbicide properties and the amount of moisture in the soil, as well as soil pH, surface area and cation exchange capacity. Soil organic matter is the principal adsorbent for many nonionic organic compounds (Hamaker and Thompson, 1972). However, there are many nonionic organic chemicals that adsorb extensively on the clay mineral fraction of soil (Yaron *et al.*, 1967). Herbicides such as glyphosate (Roundup®) can be adsorbed through formation of complexes with metal ions such as iron and aluminum. Herbicide application rates are often increased in high organic matter soils or high clay soils, or both, and decreased in sandy soils so that an effective, but safe, concentration is maintained in the soil solution.

The bonding energy between herbicides and soils is related to the type of adsorption mechanism (*e.g.* cation exchange, formation of complexes, hydrophobic interactions). As a rule, nonpolar, nonionic herbicides are adsorbed by low

Table 1. Intramolecular forces that can attract herbicides to and retain them on soil surfaces.

London-van der Waals forces:	short range bonds between the herbicide and soil that are the result of dispersion forces.
Hydrogen bonds:	bonds produced from the electrostatic attraction between an electropositive H nucleus and exposed electron pairs on electronegative atoms such as O and N.
Cation bridging:	formation of a complex between an exchangeable cation on the soil and an anionic or polar organic functional group on the herbicide.
Water bridging:	interaction between an H on an hydrating water molecule on a cation on the soil and an organic functional group on the herbicide.
Anion exchange:	electrostatic attraction of an anionic herbicide to a positively charged site on the soil surface, involving the exchange of one anion or another at the binding site.
Ligand exchange:	complex formed when a functional group on the herbicide such as carboxylate or hydroxyl displaces an inorganic hydroxyl or water molecule of a metal ion (Fe or Al) at the surface of a soil mineral.
Protonation:	Protonation of a herbicide, or formation of charge transfer complexes, at a mineral surface occurs when an organic functional group of the herbicide forms a complex with a surface proton (H).
Cation exchange:	electrostatic attraction of cationic (positively charged) organic functional groups such as amines and heterocyclic N by a negatively charged site at the soil surface.
Covalent bonding:	formation of covalent bonds between functional groups of the herbicide and the soil organic matter.

energy bonds. Adsorption of a few herbicides, such as the bipyridylium cations diquat and paraquat, involve higher energy bond formation. These herbicides are adsorbed primarily through cation exchange. However, adsorption also can involve charge transfer interactions (Kahn, 1974), hydrogen bonding and van der Waals forces (Burns et al., 1973), which have progressively lower bonding energies, respectively than cation exchange. As bonding energy increases the amount of herbicide adsorbed generally increases, so that less herbicide is in solution for plant uptake.

The soil pH can affect adsorption and influence the type of physical bonding that occurs. For instance, triazines and pyridinones are weakly basic chemicals that easily can be protonated at low soil pH levels. The pKa values (the pH at which half the molecules are protonated) for these herbicides range from about 1.7 for atrazine (AAtrex®) and fluridone (Sonar®) to 4.3 for prometon (Pramitol®). At soil pH levels less than 2 pH units above the pKa of the herbicide, adsorption is greater than in higher pH soils and there is abundant evidence for cation exchange as the bonding mechanism for triazines to soil (Carringer et al., 1975; Senesi and Testini, 1980). On the other hand, at soil pH values greater than two pH units above the pKa, more than 99 percent of the molecules of atrazine (AAtrex®) and fluridone (Sonar®) are not protonated and other mechanisms such as hydrophobic attractions, charge-transfer bonds, hydrogen bonding and van der Waals forces become more important (Hance, 1969; Hayes, 1970; Weber, 1980; Bouchard and Lavy, 1985; Weber et al., 1986). As the soil pH increases these herbicides become progressively less adsorbed to soil.

Weakly acidic herbicides such as the phenoxyacetic acids exist either as the undissociated molecule or the corresponding anion depending on the pH of the system. Adsorption of weakly acidic herbicides probably involves physical adsorption of the undissociated molecule and is not site specific. Numerous studies have shown that adsorption increases with a decrease in pH provided there is a decrease in the percent of the dissociated acid (Cheng, 1971; Koskinen et al., 1979). Other adsorption mechanisms such as charge-transfer and hydrogen honding are also possible for weakly acidic herbicides.

Observed differences in adsorption between herbicides within the same class of compounds (i.e., substituted-ureas, sulfonylureas) in the same soil are due to differences in the chemical characteristics of the herbicides. As with the soil surfaces, functional groups on an organic molecule influence the strength and mechanism of chemical retention. For example, the adsorption of phenylurea herbicides on soil increases with halogen (Cl^- or Br^-) or chlorophenoxy substitution in the phenyl ring as opposed to alkyl groups. Increasing chain length of the dialkyls or substituting the dialkyl with the corresponding alkyloxy derivative also increases adsorption (Hance, 1965; Grover, 1975).

Very little research has been conducted on adsorption of herbicide metabolites on soil. There are reports that adsorptions of some metabolites on soil can be equal to or greater than the parent herbicide (Koskinen,1984; Clay and Koskinen, 1990).

For most herbicides, a rapid, reversible equilibrium initially is established between the chemical in solution and the chemical adsorbed onto the soil surface initially. The adsorption is due to the previously described physicochemical and electrostatic forces or hydrophobic partitioning. However, once adsorbed the chemical is subject to other processes that can affect retention. Some chemicals may further react to become covalently and irreversibly bound (i.e., become

"bound residues") and some herbicides may become physically trapped within the soil particles.

Bound residue formation is a detoxification process catalyzed by soil microorganisms. Bound residues are typically defined as that portion of the residue which is not extractable with aqueous solutions or organic solvents due to incorporation of the residue into soil organic matter. Typically, radioactive carbon (^{14}C)-labeled herbicides are used in studies to quantify the nonextractable material; ^{14}C budget studies indicate that significant portions of the total applied ^{14}C-herbicide is ultimately converted to bound residue. For instance, microbial laccase and peroxidase enzymes act in coupling and polymerization reactions to bind pesticide metabolites to soil organic matter (Bollag and Loll, 1983). Chemical reactions also are important in the formation of bound residues. The binding of s-triazine residues is primarily chemical; microorganisms act only in the production of various metabolites (Bollag and Loll, 1983). Bound residues may also be formed through the uptake of herbicides or metabolites and the subsequent release of these materials through decomposition of the crop residues in the soil.

Aromatic amines (including dinitroaniline herbicide such as trifluralin [Treflan®] metabolites) can form covalent bonds with soil organic matter. Both biological and nonbiological mechanisms seem to be involved including oxidative coupling reactions of anilines to phenols and formation of anilinoquinone linkages (Hsu and Bartha, 1974a; Parris, 1980). Graveel et al. (1985) reported an initial rapid adsorption of aromatic amines, with a sharp decrease in the amount of extractable amine after 24 hours, followed by a gradual decrease over the next several weeks of incubation. This would suggest formation of covalent bonds with the soil organic matter.

There is evidence suggesting that bound or trapped forms of herbicide or metabolites may be released from organic matter. Small amounts of bound prometryn (Caparol®) and two of its metabolites were recovered from the humic and fulvic acid fractions of prometryn (Caparol®) treated soils (Khan, 1982). Helling and Krivonak (1978) found that ^{14}C-labelled bound residues of dinitroaniline herbicides were taken up by soybeans and that some of these residues were mildly phytotoxic. Dichloroaniline (DCA)-humic acid complexes (Hsu and Bartha, 1974b) and bound prometryn (Caparol®) residues (Khan and Ivarson, 1982) were degraded by microorganisms. Saxena and Bartha (1983) determined that DCA-humus complexes had faster turnover rates in soil than soil humic acid fractions, and concluded that the accumulation of DCA bound residues was unlikely. Although the release of bound residues are not expected to have economically important effects in most situations, their potential availability should not go unrecognized. The release of bound residues may be important when sensitive crops are planted after cotton.

Decreased extractability of organic chemicals with increased incubation time at times may be due to physical trapping of the herbicide within the soil particle. Decreased extractability of fluridone (Sonar®) after equilibration for 7 to 28 days

compared to one day was attributed in part to the herbicide penetrating deeper into the soil micropores and adsorbing on the interlayer surfaces of montmorillonite (Weber *et al.*, 1986). The herbicide prometon (Pramitol®) has also been shown to adsorb on the interlayer of montmorillonite (Weber *et al.*, 1965). Hydrophobic interactions and trapping of molecules in a molecular sieve formed by humic materials has been hypothesized as a retention mechanism for the incorporation of ^{14}C-prometryn into humus (Khan, 1974).

TRANSFORMATION PROCESSES

Transformation processes are those processes that actually reduce or totally eliminate the amount of herbicide present and available for plant uptake, transport and, ultimately, weed control. Herbicides are degraded by both biological and nonbiological means. Physical and chemical processes—including photodecomposition, hydrolysis, oxidation and reduction and polymerization reactions—degrade most herbicides to some extent, although the rates of these reactions in nature may be negligible for many herbicides. Nevertheless, the importance of certain physical reactions can necessitate different application strategies for specific herbicides.

The degradation of herbicides is accompanied by the formation of metabolites which, in most cases, are less toxic than the original chemicals. The metabolites of microbial degradation tend to have progressively simpler structures as herbicide degradation continues. In contrast, degradation in plants relies primarily on conjugation of residues with sugars or other compounds. The metabolites of chemical- or photo-degradation may be more novel in structure. In addition to the production of extractable metabolites, a portion of the herbicide is converted into bound residue. Bound residues typically account for a substantial portion of the applied herbicide and, as previously discussed, are formed through the chemical and enzymatic bonding of herbicides and metabolites to soil organic matter.

After reviewing reports of the degradation of sixteen herbicides in different soils, Guth (1980) suggested that most pesticides are degraded by the same pathway in different soils, although the pathways differ among pesticides. Guth also suggested that there were no differences in the pathways of degradation between laboratory and field experiments. Exceptions to this exist. For instance, long-term pesticide use may lead to the evolution of different metabolic pathways in soil microorganisms. Microorganisms which utilized *s*-triazines as nitrogen sources (Cook and Hutter, 1981) or carbon sources (Behki and Khan, 1986) were isolated from soils with histories of *s*-triazine use, but not from soils without this history. A change in the metabolic pathway of the insecticide parathion was observed in soil repeatedly treated with parathion (Sudhakar-Barik *et al.*, 1979). Additional research is needed to determine the significance of changes in microbial degradation pathways after repeated herbicide applications.

Differences in the kinetics of degradation can result in substantially different concentrations of metabolites in different soils at various times after application.

Laskowski *et al.* (1983) reviewed selected literature and determined that the variation in pesticide degradation rates in different soils was as little as 2-fold to as much as 36-fold. In all of these studies, degradation rates were measured under standard temperature and water potentials. Thus, the variation is due to the soils themselves. Although factors such as water availability, soil pH, organic matter content, microbial activity and soil aeration are known to influence each of the various degradation processes, accurate prediction of degradation rates solely from soil properties and environmental conditions is not yet possible. However, we can predict the persistence of most cotton herbicides within the degree of accuracy needed by producers for adequate weed control.

Chemical Decomposition—It has been extremely difficult to separate the individual contributions of chemical and biological processes. Experimentally, this is achieved by removing the biological component through autoclaving or by adding inhibitors of microorganisms to the soil. The risks of incomplete sterilization, contamination, or growth of resistant organisms remain as potential problems. Alternatively, chemical degradation can be demonstrated in soil-free systems where biological components can be excluded. Chemical degradation is a significant factor in the initial degradation of the chloro-*s*-triazines such as cyanazine (Bladex®) which, through hydrolysis, are converted to hydroxytriazines (Jordan *et al.*, 1970). Muir and Baker (1978) found both hydroxycyanazine and the cyanazine amide [2-chloro-4-(1-carbamoyl-1-methylethylamino)-6-ethylamino-1,3-5-triazine] in soil after cyanazine application. The cyanazine amide may be formed by nonbiological processes (Sirons *et al.*, 1973). Grayson (1986) demonstrated the potential for abiotic hydrolysis of cyanazine in weak acids. However, dipropetryn (Sancap®) and prometryn (Caparol®), both of which are methylthio-*s*-triazines used in cotton, are not converted to hydroxylated metabolites as are atrazine (AAtrex®) or cyanazine (Bladex®). These herbicides are initially converted to sulfoxides or sulfones in soil. The involvement of microorganisms in this process is unknown (Kaufman, 1974).

Photodecomposition—Photodecomposition (photolysis) is the decomposition of a herbicide induced directly by absorbing energy from light. Most herbicides have an absorption maxima in the region between 220 and 400 nm. However, only herbicides such as 2,4,5-T, chloramben (Amiben®), dinoseb and trifluralin (Treflan®) with significant absorption above 285 nm, are expected to undergo direct photolysis by sunlight. The atmosphere absorbs most UV radiation below 285 nm.

Photodecomposition of other herbicides occurs indirectly by reaction with another chemical which is activated by absorbed energy (sensitization). In experiments using artificial light at wavelengths near the herbicide's absorption maximum, a wide range of herbicides, including phenoxyalkanoic acid, *s*-triazine, substituted-urea and dinitroaniline herbicides, have been reported to

undergo photochemical reactions. These reactions have been the subject of various reviews (Plimmer, 1970; Crosby, 1976).

The contribution of photodecomposition to the persistence of many cotton herbicides is not well known. Trifluralin (Treflan®) and other dinitroanilines can be substantially decomposed by photolysis (Wright and Warren, 1966; Helling, 1976). The photocomposition of substituted-urea herbicides like diuron (Karmex®) in aqueous solutions has been observed (Hill et al., 1955; Rosen et al., 1969). In the latter studies, 69 percent of the added linuron (Lorox®) was recovered from solution following two months exposure to sunlight. The s-triazine herbicides are subject to photodecomposition under laboratory conditions (Jordan et al., 1970). Atrazine (AAtrex®) and simazine (Princep®) applied to the soil surface were degraded extensively on the sun-lit surface in comparison to the surface kept in the dark (Comes and Timmons, 1965). Small amounts of water moved fluridone (Sonar®) into the soil sufficiently to reduce but not eliminate its photodegradation in water (Jordan et al., 1970). These data suggest that fluridone may also be subject to photodegradation in soil (West et al., 1979). In contrast, perfluridone (Destun® [no longer available]) applied to soil was not susceptible to photodegradation (Ketchersid and Merkle, 1975).

The amount of herbicide decomposition by photolysis reactions in field situations is often difficult to assess. Photodecomposition is only of importance while the herbicide remains on the soil surface. A herbicide is not expected to remain on the soil surface for long however, since incorporation by diffusion and rainfall will move the majority of a herbicide some distance into the soil. Diuron (Karmex®) was partially or completely inactivated after two weeks on dry or wet soil surfaces, respectively, but the relative contributions of biodegradation, volatilization and photodecomposition were not measured (Horwitz and Herzlinger, 1974). Incorporation by irrigation increased the level of activity in these studies. Similar results were obtained by Comes and Timmons (1965). Soil incorporation of the dinitroanilines by tillage reduces photodecomposition to minimal levels.

The presence of other compounds which interact with light and the herbicide to increase or decrease photodecomposition in soils and natural waters further complicates the assessment of photodegradation. Several studies indicate that the use of silica gels or glass plates in photolysis experiments overestimates the photodegradation in comparison to soil systems (Ketchersid and Merkle, 1975; Messersmith et al., 1971; Miller and Zepp, 1983; Nilles and Zabik, 1975). Because of the complexity of natural soil systems and the lack of systematic research on many pesticides, Miller and Zepp (1983) concluded that current models of pesticide photochemistry could not predict the importance of these reactions in soil for most pesticides.

Biological Degradation—Biological degradation is the process controlling the persistence of most cotton herbicides in soils and water. Soil microorganisms reduce herbicides to nontoxic forms through a sequence of enzymatic actions on

the herbicides and their metabolites. The rates of microbial degradation are governed by biological, environmental and chemical factors. Generally, degradation results in herbicide metabolism to provide a source of carbon for microbial use. When compounds are altered but not used for growth, the process is termed cometabolism. Although initial steps in the metabolism often reduce the phytotoxicity of the herbicide, the subsequent degradation of metabolites is important also. Many herbicide metabolites are weakly phytotoxic and their accumulation in the soil could increase the risk of adverse effects to crops or the environment.

Although microorganisms which can degrade herbicides and use them as carbon sources have been isolated, other microorganisms only partially degrade herbicides. A *Bacillus sphaericus* strain degraded linuron (Lorox®), but concentrations of the metabolite, 3,4-dichloroaniline, in the culture medium increased correspondingly (Walnoffer, 1969). In soil, this metabolite would be available for degradation by other microorganisms. The $^{14}CO_2$ evolution from soil treated with [ring-^{14}C] propanil (Stam®) was increased dramatically by adding a bacterium which degraded 3,4-dichloroaniline, an initial degradation product of propanil (Zeyer and Kearney, 1982). The relative importance of such interspecies interactions is difficult to assess, but probably accounts for a significant amount of the degradation of many herbicides.

The activity of herbicide-degrading microorganisms is mediated by the soil environment and the soil physical and chemical properties. The most important of the environmental factors are soil water potential (available water content) and temperature. Decreased water availability usually results in increased persistence of the herbicide (Smith, 1985; Hitchings and Roberts, 1980; Walker, 1978). Anderson (1981) investigated the relationship between microbial biomass levels and degradation rates of diallate (Avadex®) and triallate (Far-Go®) in soils at different moisture contents. Degradation rates of both herbicides were reduced as water content decreased, but biomass levels did not decrease. Degradation rates were largely independent of biomass levels, indicating that microbial activity was reduced by the suboptimal soil water contents. Low water potentials slow the diffusion of herbicides and other substrates to the microorganisms, thereby reducing their activity (Papendick and Campbell, 1981). Bacterial activity is reduced in moderately dry soil, (water potential: -6 bars). Some fungi can remain active in extremely dry soil (water potentials as low as -30 bars) (Griffin, 1981). Even though soils may have equivalent moisture contents, differences in soil texture and microstructure may result in greatly differing levels of available water or water potentials.

Microbial activity is increased in warm soils and reduced in cold soils. In general, herbicide biodegradation also responds in this way provided that soils have adequate water content. Soil temperature effects in the field are often highly related to soil water availability. Evaporative losses tend to be greater under warmer temperatures. Temperature effects in dry soil are relatively small, due to the overriding effect of reduced soil water availability.

Other soil factors that indirectly regulate microbial degradation include soil texture, pH and organic matter levels. These factors primarily affect microorganism populations and their activities. Factors such as high nutrient availability, neutral pH, high cation exchange capacity and high organic matter levels tend to promote microbial growth and activity. Total microbial biomass or population size are only indicators of potential microbial degradation because herbicide-degrading microorganisms are only a small fraction of the total population in aerobic soils (Burge, 1969; Cullimore, 1981; Kunc and Rybarova, 1983; Loos, *et al.*, 1979). In nutrient-limited cultures, herbicides or their metabolites can be degraded to provide a source of nitrogen, sulfur or phosphorus for microorganisms (Cook *et al.*, 1978; Cook and Hutter, 1981, 1982), but the significance of these findings in the soil environment has not been established. Degradation pathways of this type might be expected in soil after addition of crop residues with high carbon to nitrogen or carbon to sulfur ratios.

Little systematic work on the effects of soil pH on herbicide degradation has been done, aside from that on the *s*-triazines (Hance, 1979). Hance (1979) found that linuron (Lorox®) degradation was increased in high pH soils, but Corbin and Upchurch (1967) found that the persistence of diuron (Karmex®) phytotoxicity was not affected by soil pH. Persistence of nitralin (Planavin®) and trifluralin (Treflan®) phytotoxicity, as determined by bioassay, was greater in acid soils than neutral soils, but quantification of the two herbicides by extraction and gas chromatography showed that only nitralin persistence was greater in acid soils (Savage, 1973). The reduced nitralin (Planavin®) degradation in acid soils was likely due to reduced microbial activity since acid hydrolysis reactions have not been reported for this herbicide in soil.

Soil aeration also greatly influences the biodegradation of herbicides. Oxygen diffuses at much greater rates in air-filled soil pores than in water-filled pores. High soil water content reduces the rate of oxygen diffusion and once the dissolved oxygen is depleted, anaerobic conditions begin to develop. Anaerobic conditions may be present in stream sediments, flooded soils, subsoils or, to a lesser extent, in microsites such as the interior of soil aggregates.

Trifluralin (Treflan®) degradation proceeded at a much greater rate in flooded soils than those kept at water contents below saturation (Probst *et al.*, 1967). Similar results were obtained in field studies with fluchloralin (Basalin®), pendimethalin (Prowl®), profluralin (Tolban®), and trifluralin (Treflan®) (Barrett and Lavy, 1983; Brewer *et al.*, 1982). The role of microorganisms as the principal agents of degradation under both aerobic and anaerobic conditions was established by comparing trifluralin degradation in autoclaved and nonautoclaved soil, and by the inhibition of trifluralin degradation by treatment with azide, a microbial inhibitor (Parr and Smith, 1973). Anaerobic dehalogenation reactions are due to microbial processes (Suflita *et al.*, 1982). Anaerobic incubations of diuron (Karmex®) and linuron (Lorox®) resulted in unusual dehalogenated metabolites (Stepp *et al.*, 1985). However, rates of herbicide degradation in the anaerobic

systems were not compared to an aerobic system in these studies. Knowledge of the pathways and relative rates of degradation for many compounds are incompletely determined or unknown.

Degradation progress curves in liquid culture and in soil are often marked by lag periods (periods of slow degradation) immediately after addition of the herbicide. Rapid degradation usually follows until the herbicide is nearly depleted. The initial lag phase is attributed to enzyme induction and synthesis or microbial adaption. Most evidence indicates that degradation enzymes are intracellular. However, acylanilide herbicides may be degraded by extracellular soil enzymes (Burns and Edwards, 1980). Several studies have shown that degradation enzymes can be induced by structurally related compounds. Walnoffer (1969) described a strain of *Bacillus sphaericus* which degraded monolinuron, linuron (Lorox®) and metobromuron, but not monuron (Telvar®), diuron (Karmex®) or fluometuron (Cotoran®). The specificity in degradation was attributed to the presence of the methoxy substituent common to monolinuron, linuron (Lorox®) and metobromuron. In later studies, the purified enzyme from this bacterium was found to degrade a wide variety of phenylamide fungicides and herbicides (Englehardt *et al.*, 1973). Chlorbromuron (Maloran®), monalide (Potablan®), propanil (Stam®), propham (IPC) and two other phenylamides induced the enzyme, but activity against linuron was less than when linuron acted as the inducer.

Other isolated enzymes also display wide ranges of specificities in their activity against structurally similar compounds. Isolated enzymes from *Pseudomonas striata* Chester, resembled those of the *Bacillus sphaericus* hydrolyzing a range of phenylcarbamate and phenylamide herbicides, respectively (Englehardt *et al.*, 1973; Kearney, 1965). The acylamidase enzymes from *Fusarium oxysporum* Schlect and *F. solani* had comparatively narrower ranges of substrates that were hydrolyzed (Blake and Kaufman, 1975; Lanzilotta and Pramer, 1970). The wide range of substrate specificities exhibited by some of these enzymes suggests that interactions between pesticides may be possible in the field. Application of herbicides with similar structures or the practice of split-applications may shorten the persistence of herbicides by reducing the lag period required for enzyme induction. The repeated application of thiocarbamate herbicides such as EPTC (Eptam®) has resulted in the accelerated biodegradation of these herbicides, and a resultant decrease in weed control. However, this phenomenon has not been demonstrated to occur with any herbicides used in cotton.

Specific structural components of the herbicide molecule can affect the enzymatic activity. The addition of sulfonate, amine or halogens to the aromatic ring tends to increase the persistence of the ring. Kearney (1967), using isolated enzyme from *Pseudomonas striata* and various phenylcarbamate substrates, found that the position and electron withdrawing potential of the ring substituents affected the rate of enzymatic hydrolysis. Strong electron withdrawing groups, such as NO_2, increased hydrolysis; the nitro group at the para position had more effect than at the meta position. Also, hydrolysis rates decreased as the size of

the side-chain increased; this was attributed to steric effects. However, with other enzymes and substrates, specific substituents may have somewhat different effects (Blake and Kaufman, 1975).

As stated earlier, persistence of herbicides is desirable while effective weed control is needed. The manipulation of persistence, either to increase or decrease the biodegradation of herbicides, is being developed to increase the effectiveness of herbicides. Application of PCMC (p-chlorophenyl methylcarbamate) with chlorpropham increased the duration of control of dodder by chlorpropham (Furloe®) (Dawson, 1969). The discovery that various carbamate and organophosphate insecticides inhibited the microbial degradation of several herbicides in soil and in pure culture led to increased interest in the manipulation of herbicide persistence (Kaufman, 1977). Dietholate, an organophosphate compound related to parathion, prolongs the persistence of several carbamothiate herbicides in soil. Further development of compounds that increase the persistence of short-lived compounds may have practical application.

HERBICIDE TRANSPORT PROCESSES

The movement of herbicides affects their potential effectiveness against weeds and the safety to crops. The performance of preemergence herbicides is often improved by the movement of the herbicide into the top few centimeters of soil. Rainfall or irrigation will accomplish this movement, or herbicide may diffuse into moist soil. Many of the dinitroaniline (Treflan®, Prowl®, others) and substituted-urea (Karmex®, Lorox®, others) herbicides used in cotton rely on retention in the surface layers of soil for selectivity with movement below this zone resulting in crop damage. Herbicide movement also is an important process in the contamination of surface and groundwater supplies. The potential contamination of groundwater and the improvements in the areas of analytical chemistry and computer modeling of chemical transport in soils are causing a reevaluation of herbicide transport in soils.

The mechanism of herbicide movement depends on the dominant exchange processes at the placement site and the physical and chemical properties of the herbicide. Herbicides in water or air will move within the medium primarily by convection in response to gravity or density. There is also short-range movement in response to concentration gradients or diffusion. The mechanism of movement also depends on the vapor pressure and water solubility of the herbicide. Soil-applied chemicals with a distribution ratio between water and air (weight/volume basis) under 10^4 (e.g., EPTC [Eptam®], trifluralin [Treflan®]) will diffuse primarily in air while those over 3×10^4 will diffuse mainly in water (e.g., diuron, s-triazines) (Goring, 1962).

Volatilization and subsequent transport of herbicides are important processes in the loss of several herbicides from soil surfaces and, in some cases, movement within the soil matrix. Vapor loss not only can reduce substantially the effective-

ness of the herbicide, but also may cause damage to cotton or non-target crops by drift. The volatility of herbicides in soil systems is related to their vapor pressures. The vapor pressures (v.p.) of herbicides range from 10^1 to 10^{-7} Pa at 68F, with most being well below 10^{-1} Pa. The greatest potential for volatility losses are herbicides that have vapor pressures greater than 10^{-2} Pa. These include all the thiocarbamates, trifluralin (Treflan®), CDAA (Randox®), dinoseb, dichlobenil (Casoron®), and some organic acid esters, among others. Herbicides with vapor pressures $< 10^{-2}$ Pa also are subject to volatility losses but to a lesser extent. For instance, volatilization of prometryn (Caparol®) (v.p. 1.3×10^{-4} Pa, 68F) from soil damaged cotton under certain conditions (Talbert *et al.*, 1971).

The magnitude of the vapor loss depends not only on the vapor pressure of the herbicide, but also on the water solubility, formulation, adsorption-desorption characteristics of soil and plant surfaces, concentration, soil water content, air flow rate, humidity, temperature and diffusion (Guenzi and Beard, 1974; Plimmer, 1976; Hance, 1980a). In general, volatility tends to increase as soil moisture or atmospheric humidity and temperature increase. As soil moisture decreases, the potential for adsorption of the herbicide on the soil increases and, therefore, the tendency toward volatilization losses decreases. As temperature increases, the vapor pressure, and hence the volatility, of a herbicide increases. For instance, prometryn (Caparol®) volatilization from the soil surface increased with increasing soil temperature and moisture (Talbert *et al.*, 1971). However, these relationships can be complicated by the fact that soil moisture can decrease as temperature increases, leading to decreases in volatility due to increased adsorption, or increases in volatility due to co-vaporization of water and herbicide.

Some herbicides can have significant movement in soil through the diffusion of volatilized herbicide. Of the herbicides used in cotton, the dinitroanilines (Treflan®, Prowl®, others) have the greatest potential for diffusion. Dinitroanilines are inherently volatile due to their high vapor pressure (Helling, 1976). Vapor transport can be decreased by decreasing the air-filled porosity of the soil (Bode *et al.*, 1973). Thus, increasing the bulk density or the soil water content tended to reduce diffusion of volatilized herbicide. However, as vapor diffusion decreased, diffusion of trifluralin (Treflan®) in the soil water increased.

The movement of herbicides in soil water depends upon rainfall or irrigation water, the macroscopic and microscopic structure of the soil, and the adsorption-desorption characteristics of the herbicide on soil. Water moves through the soil under both saturated and unsaturated conditions. When the soil is saturated with water, the pores are filled with water and transport occurs at the maximal rate. Movement of water and herbicide occurs at much slower rates under unsaturated conditions (Weber and Whitacre, 1982) because only the smaller pores are filled and water moves in response to water potential gradients. Generally, coarse-textured soils will have greater rates of water movement than fine-textured soils when saturated. However, under unsaturated conditions fine-textured soils may have greater transport rates.

In water, herbicides are transported by mass flow. Highly soluble herbicides have a greater initial potential for movement than insoluble herbicides assuming adsorption of both chemicals to soil is similar. Simultaneously, the chemical process of diffusion affects the distribution of herbicide in the water. Dispersion due to differential flow rates within pores and adsorption to soil retards the movement of herbicides relative to that of water, or a noninteracting tracer such as bromide. Soil pores have a wide range of sizes and lengths, and are highly interconnected. A portion of the pore space usually is not part of the continuous flow path or flows at much slower than average rates. Herbicides can enter and exit these spaces through diffusion. Herbicide diffusion coefficients are inversely related to adsorption, except for herbicides such as trifluralin (Treflan®) which have a high degree of movement as a volatile compound through air-filled pore space(Scott and Phillips, 1972).

Since many herbicides used in cotton are applied to the soil surface, the transport of herbicide during water infiltration is important. Water infiltration is characterized by high initial infiltration rates which decrease rapidly to a nearly constant rate. Dry soils have greater rates of infiltration than wet soils during the initial application of water. Thus, perfluridone (Destun®) movement after application of 1.5 inches of water was considerably greater in soil at a water content of <1 percent of field capacity than at 50 percent of field capacity (Ketchersid and Merkle, 1975). Other studies showed that fluometuron (Cotoran®) moved deeper into the soil in response to greater rainfall intensity or after rainfall onto a dry rather than a moist soil (Baldwin *et al.*, 1975a). In contrast, initial soil water content had little effect on the infiltration and movement of fluometuron (Cotoran®) in a very sandy soil (Wood and Davidson, 1975). The greater movement of perfluidone (Destun®) may be due to greater water solubility than fluometuron and differences in adsorption.

Adsorbed herbicides are not available for transport, but if water with lower herbicide concentration moves through the soil layer, herbicide is desorbed from the soil surface until a new equilibrium is reached. Thus, the kinetics of adsorption and desorption relative to the water conductivity rates determine the actual rate of herbicide transport. At high rates of water flow, adsorption and desorption reactions may not reach equilibrium (Hornsby and Davidson, 1973). Nonequilibrium models may describe adsorption and desorption better under these circumstances. The prediction of herbicide concentration in the soil solution is further complicated by hysteresis in the adsorption-desorption isotherms. Both adsorption and dispersion contribute to the substantial retention of herbicide found behind the initial front in typical breakthrough curves and to the depth distribution of residues.

The potential for herbicide movement is often assessed experimentally using soil columns that are treated with herbicide and leached in different ways. The herbicide is measured in the water exiting from the column or in depth increments in the column after a specific time. Another commonly-used procedure is

soil thin-layer chromatography (Helling, 1971a), where radiolabelled herbicides are chromatographed on a glass plate covered with a thin layer of soil. While these methods provide comparative estimates of mobility and have the advantage of defined conditions, many studies indicate that movement of herbicides under field conditions differs substantially from movement in laboratory soil columns (Hance et al., 1981). The potential for herbicide movement is also assessed using computer models to simulate herbicide transport as a result of specific conditions and processes.

Differences in results between laboratory and field experiments are due in part to the variabilities of rainfall, evaporation and temperature in the field. Increasing evidence also indicates that there is substantial variability within field soils (Nielson et al., 1983). This variability is due to differing physical and chemical properties of the soil within a field. Geostatistical techniques can be used to map soil properties of interest such as infiltration rate (Viera et al., 1981) or adsorption (Wood et al., 1987),and to develop sampling strategies that take field variability into account. Another difference between laboratory and field assessments is the greater degree of layering and structural influences in field soils. Soil aggregate sizes affected the distribution of herbicide in laboratory profiles following leaching (Dekkers and Barbera, 1977). This effect probably is due to the diffusion of herbicide into soil aggregates as shown by Hance (1976), but the effects of aggregates on water flow could not be discounted. Thus, the well-defined soil column is only partially representative of the soil conditions present in the field. Movement in field soils occurs over a long period of time and some herbicide is degraded during the experimental period. Laboratory studies are often conducted in much shorter time which may minimize degradation.

Herbicides used in cotton differ in their potential for downward transport in soil. Among soil-applied herbicides, the dinitroanilines appear to have little potential for downward transport (Helling, 1976). Substituted-urea and s-triazine herbicides have moderate potentials for leaching as determined by comparative movement on soil thin-layer plates (Helling, 1971b). Mobility of cotton herbicides within this group was in the order of fluometuron (Cotoran®) > prometryn (Caparol®) = diuron (Karmex®) > trifluralin (Treflan®). Dalapon (Dowpon®) was classified as highly mobile in this study. Soil thin-layer techniques also showed prometryn (Caparol®) and dipropetryn (Sancap®) to be less mobile than fluometuron (Cotoran®) (Murray et al., 1975).

Field studies with substituted-urea herbicides indicate that a small fraction of the applied herbicide moves into the subsoil. Small amounts (10-40 ppb) of diuron (Karmex®) were found at the 12-18 inch depth in cotton fields (Upchurch et al., 1969). In contrast, 10-400 ppb were found in the 0-6 and 6-12 inch depth increments. Similar results were found after repeated diuron (Karmex®) use in vineyard (Dawson et al., 1968) and orchard (Khan et al., 1976) soils. Approximately 5 ppb of diuron (Karmex®) were found in the 20-24 inch depth of the orchard soil. Small concentrations of fluometuron (Cotoran®) have also been reported in sub-

soils below the 12-inch depth (Nicholls *et al.*, 1982; Rogers *et al.*, 1985) in subsoils of cotton fields. The herbicide residues in these subsoils were not phytotoxic except to very sensitive species.

One year after a field application, 3 percent of the cyanazine (Bladex®) and 8.6 percent as the cyanazine metabolite 2-hydroxy-(1-carboxy-1-methylethylamino)-6-ethylamino-1,3,5-triazine were found at the 8-16 inch soil depth (Muir and Baker, 1978). Greater quantities of these compounds were retained in the 0.5 to 4- and 4 to 8-inch soil depths. Muir and Baker also found that another cyanazine metabolite, [2-chloro-4-(1-carbomyl-1-methylethylamino)-6-ethylamino-*s*-triazine], leached into drainage tile water in greater quantities than cyanazine. On a monthly basis, the amounts leached into the drainage water were less than 0.5 percent of that applied on a monthly basis. These results indicate that metabolites may have increased rates of transport in soil relative to the parent compound.

There is increasing evidence that some herbicides are transported deep into soil profiles at relatively rapid rates. The preferential flow of water and herbicides through macropores in soil appear to provide avenues for the rapid infiltration to considerable depths, perhaps as far as to groundwater (Simpson and Cunningham, 1982; Thomas and Phillips, 1979). Processes other than preferential flow also may be involved. Field evidence of rapid napropamide (Devrinol®) and prometryn (Caparol®) transport (Figure 2) was provided by Jury *et al.* (1986). A small portion of the napropamide (Devrinol®) and prometryn (Caparol®) moved ahead of the main bulk of the chemical which was retained at the surface. In contrast, bromide and bromacil (Hyvar®) did not have a leading peak. The lack of macropores in the nearly structureless loamy sand soil led the authors to suggest that some other transport mechanism must be involved for the napropamide (Devrinol®) and prometryn (Caparol®). The increased solubility of some pesticides in solutions of dissolved organic matter has led to speculation that herbicide-organic matter complexes which lessen the adsorptive effects of the soil might be responsible for increased rates of transport (Ballard, 1971; Khan, 1973; Madhun *et al.*, 1986). However, evidence showing appreciable levels of herbicide transport in soil by this mechanism is still lacking.

Under the right conditions or in the right situations, pesticides can be transported through soil to groundwater. A variety of pesticides have been found in the groundwater of a number of states. In one monitoring study, 17 pesticides were found in the groundwater of 33 states with concentrations ranging up to 700 ppb (Table 2) (Cohen *et al.*, 1986). Only three of the pesticides reported were herbicides used in cotton production: cyanazine (Bladex®), dinoseb and metolachlor (Dual®). Since this report, additional states have reported finding these chemicals in groundwater. Although the concentrations found are small due to the dilution of these compounds in the aquifer, it essentially is impossible to assess the total amount leached in relation to that applied.

Potential contamination of groundwater by pesticides can result from both non-point and point sources of contaminants. Non-point source contamination
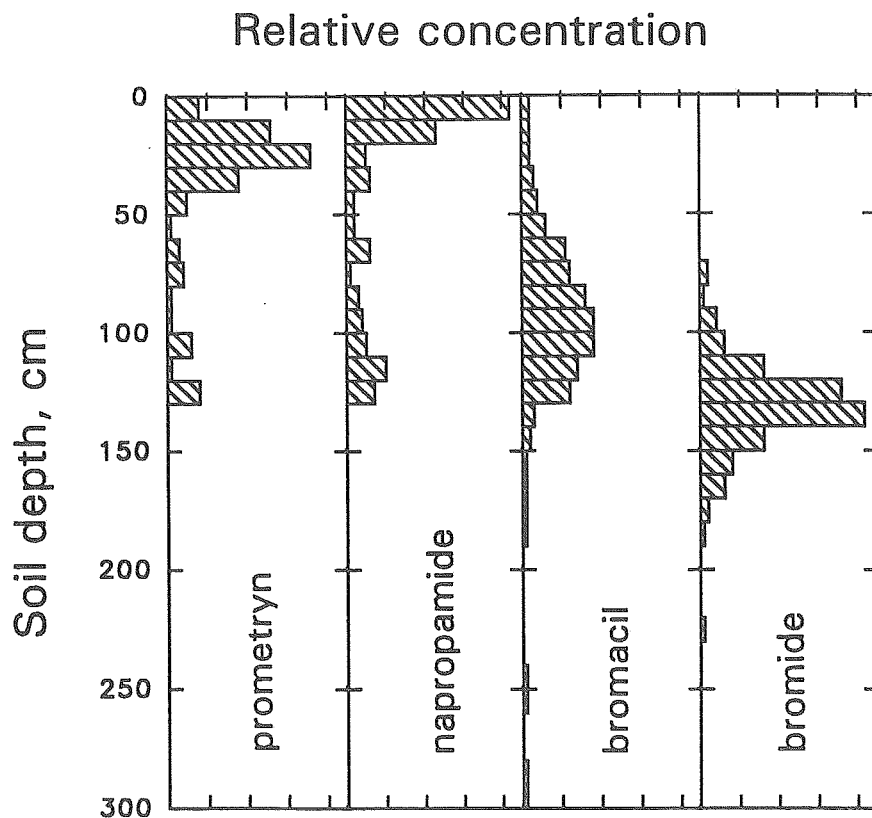
Relative concentration

Figure 2. Average relative concentrations of herbicides and bromide tracer in loamy sand field soil after leaching with 10 cm of water. (from Jury *et al.*, 1986.)

would result from movement of the pesticide through soil and subsoil following field application. This may be the avenue of entry for mobile pesiticides that are not adsorbed and are relatively persistent. Point source contamination can result from movement of pesticides through soil at pesticide disposal or mixing sites, entry through uncased or abandoned wells, or entry in surface water through natural recharge sites. Non-point *versus* point source contamination cannot be distinguished after the fact. However, isolated instances of contamination most likely would be due to point source contamination. For instance, LaFleur *et al.* (1973) reported finding fluometuron (Cotoran®), a relatively immobile chemical, in the underlying groundwater within two months after application. They obtained their water samples through an uncased well. This would indicate that the

Table 2. Pesticides Found in Groundwater of Various States. (From Cohen *et al.*, 1986.)

Pesticide	Number of states	Concentration
		(ppb)
alachlor	4	0.1-10
aldicarb	15	1-50
atrazine	5	0.3-3
bromacil	1	300
carbofuran	3	1-50
cyanazine	2	0.1-1.0
DBCP	5	0.02-20
DCPA	1	50-700
1,2-dichloropropane	4	1-50
dinoseb	1	1-5
dyfonate	1	0.1
EDB	8	0.05-20
metolachlor	2	0.1-0.4
metribuzin	1	1.0-4.3
oxamyl	2	5-65
simazine	3	0.2-3.0
1,2,3-trichloropropane	2	0.1-5.0

contamination may have been from point source rather than from non-point source contamination.

The rate of herbicide degradation in the subsoil environment will have a significant effect on the transport of herbicide over long periods of time. Several studies indicate that the rate of degradation in subsoils is substantially less than that of surface soils (Bouchard *et al.*, 1982; Harris *et al.*, 1969; Kempson-Jones and Hance, 1979; Roeth *et al.*, 1969). Longer persistence in the subsoil environment would increase the opportunity to move deeper into the subsoil. Although microorganisms have been found in surprising quantities in deep soils and aquifers, their potential activity against most herbicides is largely unknown (Ventullo and Larson, 1985). The degradation rate of 2,4-D in groundwater samples was well below that of benzoate or sodium nitrilotriacetic acid (Ventullo and Larson, 1985).

In many soils, rainfall often exceeds the infiltration rate, and water runoff occurs. Total herbicide losses in runoff from cotton fields were usually less than one percent of the applied diuron (Karmex®), linuron (Lorox®), fluometuron (Cotoran®), prometryn (Caparol®) and trifluralin (Treflan®) (Baldwin *et al.*, 1975a, 1975b; Willis *et al.*, 1975). Concentrations of herbicide in runoff waters described

for these studies ranged from levels below detection to 920 ppb, but in most instances were below 500 ppb.

Wauchope (1978) reviewed the transport of herbicides in field drainage waters and concluded that pesticide losses of herbicides formulated as wettable powders were significantly greater than those of other formulations. Losses up to 40 percent could be expected in certain situations. The factors cited by Wauchope as most important in herbicide runoff were the intensity of rainfall, length of time since pesticide application and the slope of the field. Of course, incorporated herbicides are much less susceptible to loss in runoff than surface- or foliage-applied herbicides. The factors that control infiltration of water and herbicides into the soil (soil texture and structure, water content, herbicide adsorption) also govern how much herbicide is lost in runoff. High levels of soil moisture at rainfall reduce the infiltration and increase runoff, and increase the amount of herbicide transported from the field. Under these circumstances, the amount of prometryn (Caparol®) transported in runoff reached 3 percent of the applied herbicide compared to 0.4 percent in runoff from soil that was dry at rainfall (Baldwin et al., 1975b). Herbicides in runoff are distributed in both the water and sediment according to the adsorptive capacity of the soil for the herbicide.

HERBICIDE PERSISTENCE AND ACTIVITY IN SOILS

Successful herbicide performance depends on the herbicide remaining available in sufficient quantities to be active against the weed. It has been shown that cotton produced maximum yield when kept weed-free for eight weeks after emergence (Buchanan and Burns, 1970). Therefore, to be successful, cotton herbicides must persist for this critical period, but not persist so long as to be harmful to the environment or succeeding crops. Persistence of herbicide is the net result of retention, transformation and transport processes acting in concert.

Residual levels of herbicides used in cotton may damage sensitive crops that follow cotton in the same field. The potential for damage is determined by the availability of herbicide in the soil and the sensitivity of the crop to the herbicide in question. Crops and weeds vary widely in their sensitivity to individual herbicides. In addition, prolonged periods of abnormal environmental influences may affect the microbial degradation and therefore increase or decrease herbicide persistence. The herbicide available for plant uptake is governed by the total residue concentration and the fraction adsorbed to the soil.

Organic matter is the principal soil component that must be considered in recommending herbicide application rates. In soils with a range of organic matter contents, between 0.5 and 3.0 lb. per acre of trifluralin (Treflan®) were required for 80 percent weed control four weeks after planting (Weber et al., 1987). There was a high correlation between the amount of herbicide required for weed control and organic matter content. Similar results were obtained with metolachlor (Dual®). Increasing application rates above recommended levels increases the

risk of carryover, while decreasing herbicide rates may reduce the period of control obtained for some weeds.

Most herbicides used in cotton are dissipated by the previously mentioned degradation processes during the first year after application. Therefore, carryover and subsequent accumulation would be limited. If a herbicide were applied at 1.0 pound per acre and 10 percent of the chemical carried over, in the fifth year the maximum amount of chemical present would be 1.11 pounds per acre. At 20 percent carryover, there would be 1.25 pounds per acre the fifth year (Table 3).

Table 3. Accumulation of herbicide applied at 1.0 pound per acre with 10 and 20 percent carryover.

Year	10 percent carryover		20 percent carryover	
	Carryover	Total	Carryover	Total
	(lb/A)			
1	0	1.00	0	1.00
2	0.10	1.10	0.20	1.20
3	0.11	1.11	0.24	1.24
4	0.11	1.11	0.25	1.25
5	0.11	1.11	0.25	1.25

Parka and Tepe (1969) found that between 0.6 and 30 percent of the trifluralin (Treflan®) remained five to seven months after application, but in most soils the range was from 2-6 percent. After applications of 24 and 15 pounds of trifluralin over four- and three-year periods, respectively, herbicide levels were not over 4.1 percent (0.98 ppm), indicating that trifluralin does not accumulate in soils. Numerous other studies support the conclusion that trifluralin and other dinitroaniline herbicides do not accumulate in soils suitable for cotton (Miller et al., 1975; Golab et al., 1979; Brewer et al., 1982). In soils of more northern areas, where cold temperatures slow degradation, trifluralin is more persistent, and significant amounts of herbicide can carry over at high rates of application (Solbakken, et al., 1982; Smith and Muir, 1984).

Trifluralin (Treflan®) residues may affect the growth of rotational crops following cotton. Stoller and Wax (1977) found that dinitroaniline herbicides were sufficiently dissipated after 5 and 12 months to allow the normal growth of wheat and corn, respectively. Miller et al. (1975) found that trifluralin damaged grain sorghum and Japanese millet 15 months after application. The soil concentrations of trifluralin at 15 months were approximately 1 ppmw in the top 12 inches of soil. Similar effects were found with nitralin (Planavin®) and benefin (Balan®). In other studies, trifluralin damage to oats was dependent on the application rate (Schwiezer and Holstun, 1966). Little damage was observed at the 1 pound per

acre rate, but oats were damaged at the 2 and 4 pounds per acre rates 6 months after application.

The degradation of dinitroaniline herbicides results in a large number of metabolites similar in structure to the parent chemical (Probst et al., 1967; Kearney et al., 1976; Golab et al., 1975). A simplified scheme for the degradation of the dinitroanilines is shown in Figure 3. Further details of the metabolism and fate of these herbicides can be found in previous reviews (Helling, 1976; Kaufman, 1974). Long term use of trifluralin (Treflan®) has the potential for accumulation of metabolites to levels that could have adverse effects on cotton production. The metabolites of trifluralin (Treflan®) have been shown, however, to be nonphytotoxic to cotton, even at relatively high concentrations (Koskinen et al., 1984; Koskinen et al., 1985; Vaughn and Koskinen, 1987). Boyette et al. (1988) found only small effects by trifluralin metabolites on microbial decomposition processes at rates of chemicals much higher than would be found in field situations.

Substituted-urea herbicide persistence has also been extensively studied. Typically between 75 and 90 percent of the applied diuron (Karmex®) or linuron (Lorox®) is dissipated in the first year after field application (Chandler and Savage, 1980; Hill et al., 1955; Khan et al., 1976; Upchurch et al., 1969). Related studies showed that eight months after application diuron (Karmex®) was not present in high enough concentrations to damage cover crops (Hill et al., 1955). Although small amounts of diuron (Karmex®) and the metabolite 3,4-dichloroaniline can carry over, there is no indication that the residues accumulate in soils repeatedly treated with diuron (Khan et al., 1976). Fluometuron (Cotoran®) has the potential to carry over in sufficient quantities to damage some rotational crops after use in cotton, but does not accumulate in appreciable quantities (Chandler and Savage, 1980; Rogers et al., 1985; Rogers et al., 1986). Application of the insecticide aldicarb (Temik®) reduced the rate of degradation of fluometuron (Cotoran®) and dinitramine (Cobex®), but both herbicides had less than 5 percent of applied remaining after 120 days (Gomaa et al., 1979).

The degradation of fluometuron (Cotoran®) and diuron (Karmex®) by several pathways leads to the formation of substituted aniline metabolites (Figure 4) similar in structure to the parent chemical. These metabolites have been shown not to affect growth and yield of cotton even at concentrations much higher than would be found in normal field situations (Moorman and Koskinen, 1990).

The s-triazines principally used in cotton (cyanazine [Bladex®], prometryn [Caparol®], dipropetryn [Sancap®]) are of short to moderate persistence in soil. Cyanazine (Bladex®) half-lives were estimated to be from 11 to 24 days over a 3-year field study (Muir and Baker, 1978). In another field study, only traces of cyanazine (Bladex®) remained after 5 months (Sirons et al., 1973). From 2 to 23 percent of the applied prometryn (Caparol®) remained in soil 7 months after the last of three annual applications (Weber et al., 1974). In this study, the greater
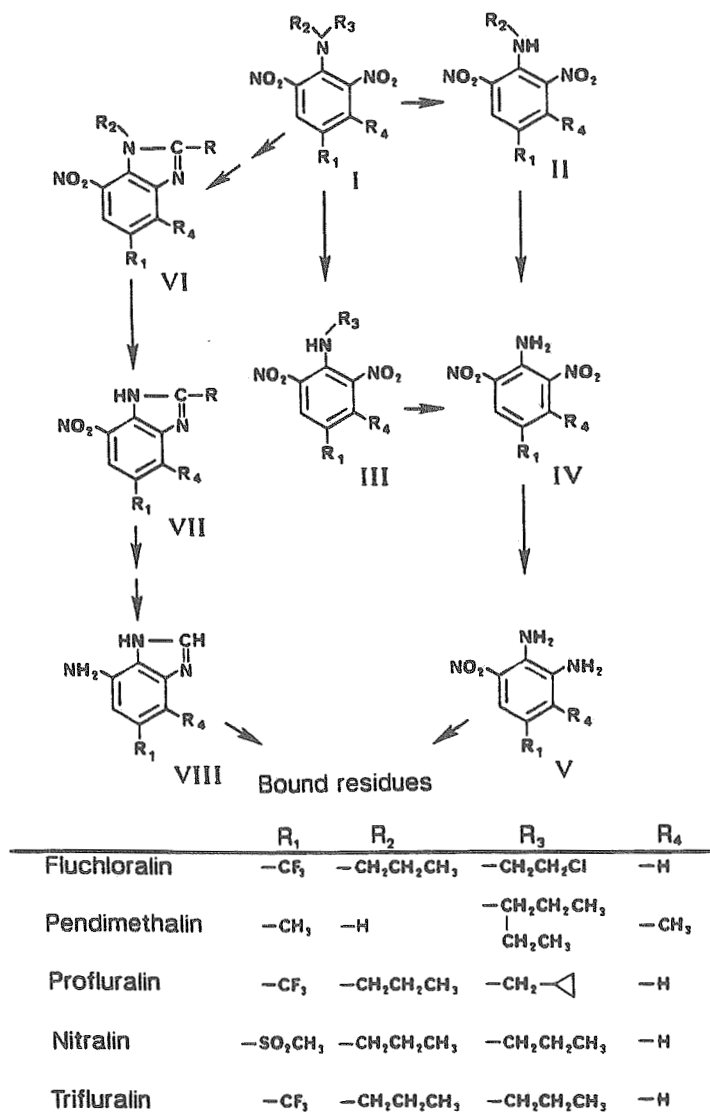
	R_1	R_2	R_3	R_4
Fluchloralin	$-CF_3$	$-CH_2CH_2CH_3$	$-CH_2CH_2Cl$	$-H$
Pendimethalin	$-CH_3$	$-H$	$-CH_2CH_2CH_3$ / CH_2CH_3	$-CH_3$
Profluralin	$-CF_3$	$-CH_2CH_2CH_3$	$-CH_2-\triangleleft$	$-H$
Nitralin	$-SO_2CH_3$	$-CH_2CH_2CH_3$	$-CH_2CH_2CH_3$	$-H$
Trifluralin	$-CF_3$	$-CH_2CH_2CH_3$	$-CH_2CH_2CH_3$	$-H$

Figure 3. Probable pathways of dinitroaniline degradation in aerobic soils. (From Golab *et al.*, 1975; Golab *et al.*, 1979; Kearney *et al.*, 1976; Laanio *et al.*, 1973; Stralka and Camper, 1981.)

residue levels were in the soils amended with massive amounts of clay or organic matter. Persistence or carryover problems with these herbicides applied in cotton are not anticipated.
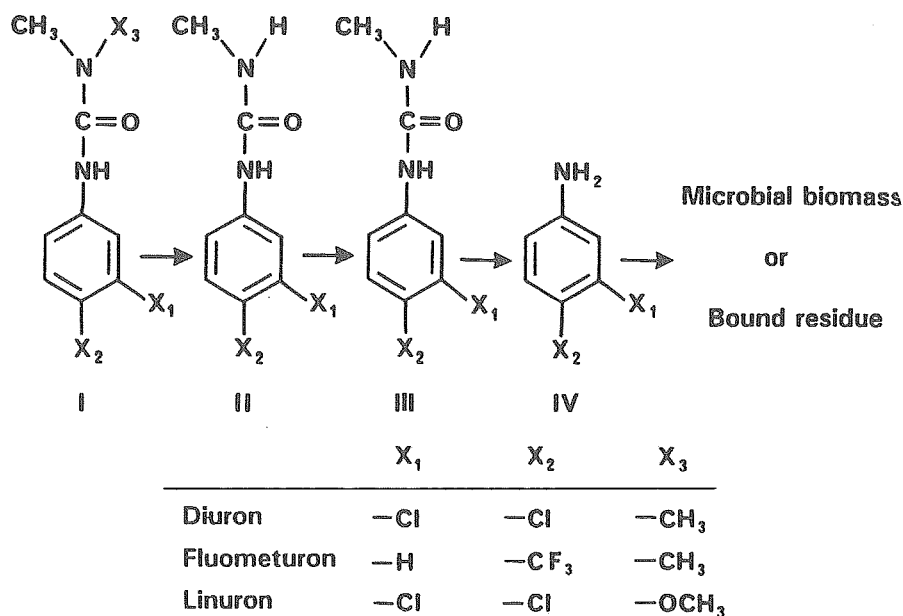
	X_1	X_2	X_3
Diuron	$-Cl$	$-Cl$	$-CH_3$
Fluometuron	$-H$	$-CF_3$	$-CH_3$
Linuron	$-Cl$	$-Cl$	$-OCH_3$

Figure 4. Pathway of diuron, fluometuron, and linuron degradation in aerobic soils. (From Bozarth and Funderburk, 1971; Dalton et al., 1966; Ross and Tweedy, 1970; Tweedy et al., 1970; Wallnofer, 1969.)

Some polycyclic phenoxyalkanoic acid (PCA) herbicides are used or have potential for use in cotton (e.g. fluazifop-butyl [Fusilade®], oxyfluorfen [Goal®], lactofen [Cobra®]). PCA herbicides are added to soil as esters that are rapidly hydrolyzed to herbicidally active acids (Martens, 1978; Bewick, 1986). The subsequent degradation of the acids occurs at slower rates, with the aliphatic side chains being degraded before the aromatic rings (Martens, 1978; Smith, 1985). The acids formed from these compounds are moderately persistent in soil, with 50 percent being degraded in 50 days or less. Limited evidence suggests that these herbicides persist longer in low pH soils (Hance, 1979). This would be consistent with the expected effect of low pH on the ester hydrolysis reaction.

Limited information is available on the field persistence of other herbicides that can be used in cotton. Norflurazon (Zorial®) was present in five field soils in quantities sufficient to injure grain sorghum (Sorghum bicolor L. Moench) one year after application (Schroeder and Banks, 1986a). Fluridone (Sonar®) persistence in these same soils was less than one year and injury to grain sorghum was not observed the year following application (Schroeder and Banks, 1986b). Methazole (Probe®) is rapidly degraded in soils to 1-(3,4-dichlorophenyl)-3-meth-

ylurea which is moderately persistent and contributes to the residual herbicidal activity of this compound (Walker, 1978).

INTERACTIONS OF SOIL FACTORS/PROCESSES AND CULTURAL PRACTICES AND HERBICIDE ACTIVITY

TILLAGE OPERATIONS

As previously discussed, herbicide persistence and efficacy in soil are controlled by various soil processes including adsorption, decomposition and movement. These processes are directly or indirectly affected by soil properties such as soil moisture, temperature, pH, organic matter and microbial populations. Tillage systems affect these same soil properties (Blevins et al., 1983; Phillips and Phillips, 1984; Wiese, 1985; Koskinen and McWhorter, 1986).

Conventional tillage systems can decrease the soil moisture compared to conservation tillage systems. This is primarily due to increased runoff and decreased infiltration. The first-stage evaporation also increases (Phillips, 1984; Weber and Lowder, 1985). The decreased moisture in conventional tillage systems has the potential to decrease the degradation, volatilization and leaching of the herbicide.

Different tillage systems have different effects on soil temperature. In general, increased tillage increases the soil temperature in the spring and summer. The magnitude of the increase depends on the geographical location and the soil depth at which the temperature is measured. The rate of chemical reactions and microbial processes are temperature dependent, therefore, increases in soil temperature can lead to increased decomposition. Increased temperature also increases the volatilization of the herbicide.

In some situations, continuous application of ammonium fertilizer in conservation tillage systems decreases soil pH substantially compared to conventional tillage systems. Lack of tillage causes ammonium fertilizer to be nitrified near the soil surface. This lowers the soil pH in the vicinity of the fertilizer. With tillage, the fertilizer is mixed in the soil, diluting the pH effects. The low pH around the nitrifying fertilizer affects herbicide adsorption and chemical and microbial decomposition as previously discussed.

Tillage dramatically affects the organic matter content of soil. Once a soil is plowed for the first time, the organic matter begins to decrease. Each tillage operation accelerates the organic matter decomposition, particularly in warm moist climates. Finally, an equilibrium value is reached where the inputs equal the degradation. Most cotton producing areas are at this low equilibrium level.

Leaving residue on the surface, as in conservation tillage, or plowing under cover crops will start the process of restoring the organic matter content. With increased organic matter, there will be increased herbicide adsorption and de-

creased availability. Crop residues on the soil surface intercept and retain herbicides—in some cases more than 50 percent of that applied (Banks and Robinson, 1982).

Soil microbial populations can be substantially lower in soil under conventional tillage systems than under conservation tillage systems. The differences are primarily confined to the top three inches of soil (Doran, 1980; Dick, 1984). The differences in microbial populations, activities and ecology between tillage systems are primarily the result of differences in soil moisture, pH, temperature and organic matter content.

As discussed, tillage systems affect various soil properties which, in turn, can affect herbicide persistence. Many studies report greater persistence of triazine herbicides (AAtrex®, Caparol®, others) in soil under conventional tillage systems compared to no-till systems (Witt, 1984). However, if a residue does remain at the end of a growing season, there is more risk to the rotational crop planted with no-till practices rather than with conventional tillage because the herbicide residue is left in a shallow concentrated band.

Tillage and seedbed preparations can also affect the distribution of herbicides in soil. For instance, trifluralin (Treflan®) is preplant incorporated resulting in a uniform distribution of herbicide in the surface two to three inches of soil. In the bedding process, soil is removed and raised beds are constructed. Even after knocking down the beds, the depth of herbicide treated soil is greater in the row than in the furrow.

APPLICATION METHODS

For herbicides that are subject to losses due to volatilization and photodecomposition, or both, incorporation of the herbicide can increase its persistence. Soil incorporation has been shown to dramatically reduce the losses of trifluralin (Treflan®) and increase its efficacy. Increase in persistence has been found within the depth of incorporation (Savage and Barrentine, 1969; White et al., 1977). However, incorporation must be as soon after application as possible, e.g., within 24 hours for trifluralin (Treflan®). Increased persistence with soil incorporation has been demonstrated for prometryn (Caparol®) (Walker, 1976), diuron (Karmex®) and fluometuron (Cotoran®) (Horowitz and Herzlinger, 1974).

Another method for reducing volatilization, photodecomposition and even leaching losses is to formulate and apply herbicides as granules. Volatile herbicides such as triallate (Far-Go®), EPTC (Eptam®), dichlobenil (Casoron®) and propham (Chem-Hoe®) are normally incorporated into soil to avoid rapid loss. These herbicides have been shown to be more persistent when formulated as granules that could be surface-applied without incorporation, as opposed to other formulations (Parochetti et al., 1971; Hance et al., 1973; Siegman et al., 1975; Schreiber, 1976; Spencer-Jones and Wilson, 1976). Prometryn (Caparol®) applied to soil as granules reduced volatilization and subsequent injury to cotton compared to prometryn applied as emulsifiable concentrate or wettable powder

formulations (Talbert *et al.*, 1971). Based on a literature review, it was concluded (Edwards, 1972) that for insecticides the relative order of persistence was granules > emulsions > miscible liquids > wettable powders. In general, granular herbicides are also more persistent than other formulations.

Herbicides have been applied through different irrigation systems (drip, furrow and sprinkler irrigation) with varying degrees of success. Herbicides with water solubilities greater than 0.1 mM have worked the best. Alachlor (Lasso®), atrazine (AAtrex®), metribuzin (Sencor,® Lexone®), napropamide (Devrinol®), and various thiocarbamates have all been used effectively in irrigation systems (Combellack, 1984). Trifluralin (Treflan®) has been reported to inadequately control weeds when applied and incorporated by high pressure sprinkler irrigation systems (Smith *et al.*, 1973; Ogg, 1980), apparently due to low solubility (< 1.0 μM) in water, strong adsorption to soil, volatilization losses or a combination of the three. However, new sprinkler systems with low pressure drop nozzles may give acceptable weed control with trifluralin (Treflan®) and pendimethalin (Prowl®).

Although good weed control has been reported using a center pivot sprinkler system to apply herbicides, the method has potential problems with certain types of herbicides. Volatilization losses during center pivot application can be large for herbicides with high vapor pressures and relatively low water solubilities. Also, herbicides formulated as wettable powders have not been applied through sprinklers because problems occur when they settle out of solution and clog sprinkler lines.

SUMMARY

The performance and persistence of cotton herbicides is not a fixed property of the herbicide. It is regulated by the interaction of pesticide, soil, plant and environmental factors with transformation, retention and transport processes. Pesticide factors include: chemical properties such as structure, solubility and volatility; application methods including formulation and rate; and decomposition characteristics such as mechanisms, kinetics and metabolite formulation. Important soil factors include organic matter and clay type and amount, pH, moisture, aeration, microbial activity, topography and depth to groundwater. The major plant factor is the species characteristics combined with stage of growth. Environmental factors include temperature, precipitation, air movement and solar radiation.

While many of these factors vary across the cotton producing regions and season, they can also vary substantially from field to field within a farm and from day to day. It would appear that only rare combinations of factors and their effects on processes would result in performance and persistence problems such as carryover, lack of weed control or environmental contamination. For instance, most of the herbicides applied for preemergence weed control in cotton

have created no particular carryover problems. In the West there have been some persistence problems where rainfall has been critically short. In the Mid-South and Southeast, some persistence problems have resulted from the use of herbicides such as diuron (Karmex®) and linuron (Lorox®) applied at layby, especially where rainfall was critcially short late in the growing season, thus delaying degradation of the herbicide.

Lack of weed control with repeated herbicide applications, similar to problems associated with thiocarbamate herbicides in corn, have not been observed in cotton production. It would also appear that groundwater contamination by herbicides would not be a problem in cotton production. No incidences of non-point pollution have been reported.

There are no apparent persistence problems with some of the new chemistries such as the monophenyl- and diphenyl-ether sethoxydin (Poast®) and fluazifop (Fusilade®) type herbicides that are applied broadcast over-the-top for selective control of grasses. However, as new chemicals are introduced and used for weed control in cotton production, knowledge of factors and processes and their interactions and how they affect herbicide persistence in soil will prevent future potential problems. The knowledge also will aid in the future development of crop rotation systems for cotton.

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