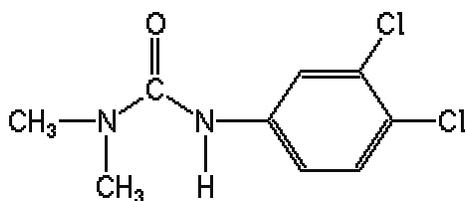


## ENVIRONMENTAL FATE OF DIURON

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This document reviews the environmental fate of diuron. The chemical (IUPAC) name for diuron is N-(3, 4-dichlorophenyl)-N, N-dimethylurea (C<sub>9</sub>H<sub>10</sub>Cl<sub>2</sub>N<sub>2</sub>O). In California, registered products containing the active ingredient diuron include Direx, Karmex, and Krovar (in combination with bromacil), among others. Diuron is often used in combination with other herbicides such as bromacil, hexazinone, paraquat, thiadiazuron, imazapyr, monosodium, sodium chlorate, sodium metaborate, and copper sulfate (U.S. EPA, 2004a).



Diuron

### General Information and Mode of Action

Diuron is a systemic substituted phenylurea herbicide. Diuron is easily taken up from soil solution by the root system of plants and rapidly translocated into stems and leaves by the transpiration system, moving primarily via the xylem. Diuron primarily functions by inhibiting the Hill reaction in photosynthesis, limiting the production of high-energy compounds such as adenosine triphosphate (ATP) used for various metabolic processes. Diuron binds to the Q<sub>B</sub>-binding niche on D1 protein of the photosystem II complex in chloroplast thylakoid membranes, thus blocking electron transport from Q<sub>A</sub> to Q<sub>B</sub>. This process prevents CO<sub>2</sub> fixation and the production of ATP and other high energy compounds which are needed for plant growth. The inability to reoxidize Q<sub>A</sub> promotes the formation of triplet state chlorophyll, which interacts with ground state oxygen to form singlet oxygen. Both triplet chlorophyll and singlet oxygen can extract hydrogen from unsaturated lipids, producing a lipid radical and initiating a chain reaction of lipid peroxidation. Lipids and proteins are attacked and oxidized, resulting in loss of chlorophyll and carotenoids, and in leaky membranes which cause cells and cell organelles to dry and disintegrate rapidly (Hess and Warren, 2002).

Diuron is available in wettable powder, granular, flowable, pelleted/tableted, liquid suspension, and soluble concentrate formulations. Technical diuron is a white, crystalline, and odorless solid. Diuron is stable in neutral media at normal temperatures, and is hydrolyzed by acid and alkalis. It is stable towards oxidation and moisture under normal conditions and decomposes at 180-190 °C (Helliwell et al., 1998).

Diuron is a broad-spectrum residual herbicide registered for preemergent and postemergent control of both broadleaf and annual grassy weeds. When diuron is used on preemergent weeds, it allows seeds to germinate normally, but causes them to lose their green color, after which they soon die of starvation (Ferrell et al., 2004). In the U.S. diuron is used on a variety of fruit and nut crops, grains, cotton, corn, sorghum, mint, gum, asparagus, sugarcane, seed crops, coffee, hay, cut flowers, and for fallow and idle cropland use. It may be used in irrigation and drainage systems when water is not present. Diuron also has widespread use in non-agricultural applications, especially industrial and rights of way uses, where often in combination with other herbicides it provides total vegetation control. These applications include along fence lines, pipelines, powerlines, railway lines, roads, footpaths; in timber yards and storage areas; and around commercial, industrial and farm buildings, electrical substations, and petroleum storage tanks. It has some use as an algaecide in ornamental ponds, fountains, and aquaria, but not natural water bodies. It may be used as a mildewicide in paints used on buildings and structures. A former use as an antifouling paint is no longer registered in the U.S., but may be registered in other countries (U.S. EPA, 2004a).

Diuron is one of the most commonly used pesticides in California. Reported statewide use in 2002 was 1,303,745 pounds. Of this reported use, 48 percent was applied to rights of way, followed by 18 percent and 14 percent on alfalfa and oranges, respectively. Additional 2002 uses in California included grapes (4.3 percent), landscape maintenance (3.7 percent), walnuts (2.6 percent), and cotton (1.6 percent).

**Table 1. Physical-Chemical Properties**

Molecular weight <sup>a</sup>	233.10 g/mole
Water solubility <sup>a</sup>	36.4 ppm (25° C)
Vapor pressure <sup>a</sup>	6.90 x10 <sup>-8</sup> mm Hg (25° C)
Hydrolysis half-life <sup>a</sup>	1490 days (pH 5) 1240-1330 days (pH 7) 2020 days (pH 9)
Aqueous photolysis half-life <sup>a</sup>	43.1–2180 days (pH 7 at 25° C)
Aerobic soil degradation <sup>a</sup>	372 days
Anaerobic soil degradation <sup>a</sup>	995 days
Soil photolysis half-life <sup>a</sup>	173 days
Field dissipation half-life <sup>a</sup>	99.9-134 days
Henry's law constant <sup>a</sup>	5.10 x 10 <sup>-10</sup> atm m <sup>3</sup> mol <sup>-1</sup> (25° C)
Octanol-water coefficient: Kow <sup>a</sup>	648 - 747
Soil adsorption coefficient: Koc <sup>b</sup>	418-560

<sup>a</sup> DPR Pesticide Chemistry Database, 2003.

<sup>b</sup> ARS USDA, 2004

**Table 2. Ecological Toxicity<sup>c</sup>**

Mallard Duck LD50	>2000mg/kg
Mallard Duck 8 day LC50	>5000 ppm
Bobwhite Quail 8 day LC50	1730 ppm
Bluegill Sunfish LC50 (96 hrs)	5.9 ppm
Rainbow Trout LC50 (96 hrs)	190 ppm
Sheepshead Minnow LC50 (96 hrs)	6.7 ppm
Daphnia Magna LC50 (48 hrs)	8 ppm
Honeybee LD50 (48 hrs)	145 ug/bee
Oyster Shell EC50 (96 hrs)	4.8 ppm

<sup>c</sup> USEPA, 2004b

## **AIR**

Diuron is applied by broadcast or band spray on soil surface using ground or aerial equipment, suggesting that there is a possibility of drift. However, diuron is non-volatile, as indicated by its low vapor pressure of  $6.90 \times 10^{-8}$  mm Hg (25° C), and a low Henry's law constant of  $5.10 \times 10^{-10}$  atm m<sup>3</sup> mol<sup>-1</sup>. Its low vapor pressure and low Henry's law constant indicate that diuron is unlikely to be dispersed in air over a large area and has a low tendency to volatilize from water or moist soils. Volatilization is insignificant except when diuron is exposed on the soil surface for several days or weeks under hot, dry conditions (Hess and Warren, 2002).

## **WATER**

Diuron's relatively low  $K_{OC}$  indicates a relatively low tendency to sorb to soils and sediments, while its hydrolysis and aqueous photolysis half-lives are relatively long. Consequently diuron is both mobile and relatively persistent, and is therefore prone to off-site movement in surface runoff, and migration to ground water.

Andrieux et al. (1997) assessed losses of diuron via runoff from vineyards under natural Mediterranean climate conditions. During the growing season of 1994 and 1995, runoff and diuron concentrations were monitored at two field sites, one tilled and one under no-till management. Despite a time lag of greater than 4 months between application and the first runoff event in 1994, diuron concentrations in overland flow exceeded 200 ug/L at the no-till in both years of investigations. In 1995, the first strong rainfall following application removed 60% of total diuron runoff loss at both sites, although it accounted for 17 and 7% of the total seasonal runoff volume at the no-till and tilled site, respectively. In 1995, seasonal diuron loss at the no-till sites was 1.71 % and at the tilled site was 0.68%. No herbicide residues were detected in soil samples collected 1 week prior to chemical spraying in 1994 and 1995. The authors suggested that moderate temperatures and moisture conditions during winter facilitated complete microbial decomposition. In four-months of soil sampling following herbicide treatment there were no detections below a soil depth of 2 cm at the no-till plot. However, at the tilled site, pesticides were present in samples at depths of 0-15 cm 17 days after spraying to three months. The authors suggested that infiltration increases attributable to tillage effects accounted for diuron distribution in the soil profile. Andrieux et al. (1997) concluded that the persistence of diuron

coupled with tillage may provide the potential for leaching.

In contrast, no-till practices have been shown to contribute to ground water contamination under certain California conditions. Braun and Hawkins (1991) conducted a rainfall runoff monitoring study in a citrus-growing region of Tulare County, California, where growers commonly avoid cultivation of citrus row middles. The row middles under such a management practice are typically compacted, with low infiltration rates and a high tendency to yield runoff during storm events. One method of disposing of excess runoff water in the area is to utilize dry wells that allow drainage of runoff through the shallow hard-pan soil layer. This mechanism allows direct introduction of residues to the subsurface and subsequent potential migration to shallow ground water. Although diuron applications had occurred at least 2 months prior to sampling, relatively high concentrations of diuron in runoff water entering dry wells were found, ranging up to 890  $\mu\text{g L}^{-1}$ . The data provide strong evidence that the widespread regional presence of simazine and diuron in ground water is at least partially attributable to contaminated runoff water entering dry wells.

Spurlock et al. (1997) examined the runoff of diuron, simazine, and bromacil from citrus orchard middles under simulated rainfall conditions. Peak diuron runoff concentrations were comparable to those reported by Braun and Hawkins (1991), ranging from 600 to 1700  $\mu\text{g L}^{-1}$ . Little difference in runoff concentrations of the three preemergent herbicides was observed indicating that herbicide substitution would be an ineffective mitigation measure. They concluded that low soil permeability arising from hardpan layers or compaction drives the runoff process. These same factors are probably responsible for diuron off-site movement in runoff water from rights-of-way because rights-of-way are generally engineered to maximize water run off.

Powell et al. (1996) studied off-site movement of diuron in surface water from a highway shoulder right-of-way application. Diuron was applied in a spray to a 2.4-meter wide strip next to the highway pavement, at a rate of 3.59  $\text{kg diuron ha}^{-1}$ . Simulated rain (13 mm in 1 hr) was applied to plots on treated highway shoulders at three sites. Diuron was detected at two of three sites in the runoff of one hour of simulated rainfall from highway shoulder plots. Concentrations of diuron were as high as 1770  $\mu\text{g L}^{-1}$  in runoff (combined water and sediment), from sampling conducted one day after herbicide application. From 0.2 to 5.4% of the diuron applied to highway shoulders moved off-site in runoff during the simulations. Simulated rain was applied to treated highway shoulder at intervals of 0, 2, and 4 wk after herbicide application. The highest percentages were observed when rain was simulated 1 day after herbicide application.

Diuron is typically applied in winter or early spring during California's rainy season to control weeds. This practice, coupled with relatively high use, is one reason why diuron is one of the most commonly detected pesticides in California surface water. Diuron was detected in more than half of 955 surface water samples analyzed for diuron and reported in DPR's surface water database. Typical reporting limits for the diuron analyses are 0.05 - 0.10  $\mu\text{g}^{-1}$ . While detected concentrations range from 0.01 to 30.6  $\mu\text{g L}^{-1}$ , the majority of concentrations range from 0.1 to 1  $\mu\text{g L}^{-1}$ . These concentrations were approximately equal to the 20<sup>th</sup> and 80<sup>th</sup> percentiles of detected concentrations. The highest concentrations and most frequent detections occur during December-March, which coincides with both the rainy season and the peak diuron application season.

Microbes are the primary agents in the degradation of diuron in aquatic environments. The aerobic biodegradation pathway for diuron is well established (Figure 1), proceeding by successive demethylation steps to form DCPMU, DCPU [1-(3,4-dichlorophenyl)urea] and DCA (3,4-dichloroaniline). Reductive dechlorination has been observed in anaerobic pond sediments and leads to the formation of the dechlorinated product, 3-(3-chlorophenyl)-1,1 dimethylurea (Field et al., 1997).

Boule et al. (1997) reported that the major photoproducts observed in the photolysis of diuron [3-(3,4-dichlorophenyl)-1,1-dimethylurea] in aqueous solution resulted from a heterolytic substitution of chlorine by OH (Figure 2). A wavelength effect was observed: at 254 nm the formation of 3-(4-chloro-3-hydroxyphenyl)-1,1-dimethylurea accounted for more than 90% of the conversion, whereas when the solution was irradiated in 'black light' (85% of photons emitted at 365 nm, about 7% at 344 nm), the major photoproduct was 3-(3-chloro-4-hydroxyphenyl)-1,1-dimethylurea. However, as noted previously, photolysis is not generally a principal route of diuron degradation in aqueous systems.

## SOIL

Diuron is moderately to highly persistent in soils. The commonly reported average field dissipation half-life is 90 days, although such half-lives are typically highly variable. Phytotoxic residues generally dissipate within a season when applied at low selective rates. At higher application rates, residues may persist for more than one year (Kidd and James, 1991). Microbial degradation is the primary means of diuron dissipation from soil. Photodegradation is not considered a primary dissipation route, but losses can be significant if diuron remains on the soil surface for several days or weeks (Hess and Warren, 2002).

Diuron is mobile in soil. Similar to many other pesticides, diuron sorption is highly correlated with organic matter (Spurlock and Biggar, 1994). Consequently leaching is greatest in low organic matter soils. Other soil conditions that favor diuron leaching include high soil permeability to water, such as in coarse soils.

Due to the diuron's persistence and mobility, the herbicide is one of the most commonly detected currently registered pesticides in California's ground water (Troiano et al., 2001). As of June 2004, there were confirmed detections of diuron in 418 California water wells reported in DPR's well inventory database (WIDB). Typical diuron reporting limits in the WIDB are  $0.05 \text{ ug L}^{-1}$ , and detected diuron concentrations range up to  $3.96 \text{ ug L}^{-1}$ . Diuron is considered a known California ground water contaminant, and its use is regulated under DPR's ground water protection regulations online at <http://www.cdpr.ca.gov>.

In the rights-of-way runoff study of Powell et al. (1996) discussed previously, post-simulation and end of season soil cores at the two sites with measurable runoff had detectable diuron residues only in the top 15 cm (0.5 ft). However, cores taken prior to application had diuron down to 0.30 m, presumably from the past year's application (Powell et al., 1996). At a third simulated rainfall site, no runoff left the plots. The absence of detectable residues in soil to a depth of 3.0 m (10 ft) suggests that much of the herbicide applied to the shoulder may have leached rapidly through the coarse gravelly soil (Powell et al., 1996). The soil data was insufficient to yield definite conclusions about leaching in infiltration areas. However, the levels

of diuron found in runoff suggest that dry wells, if present, would provide an important conduit for the transport of diuron to groundwater. The authors concluded that rights-of-ways with high percentages of gravel and sand in the soil may be of concern, since diuron may be transported rapidly to depths greater than 0.3 m (Powell et al., 1996).

Bulcke et al. (2000) evaluated soil-applied diuron degradation in a pear orchard planted on loam soil after treatment with 3 kg ai ha<sup>-1</sup> of diuron. In plots treated with the same herbicide for 12 years, the soil half-life was 37 days. In other plots that hadn't been previously treated the rate of diuron soil dissipation was 2.2 times lower, indicating moderately enhanced biodegradation in the previously treated plots. Herbicide biodegradation is defined as enhanced when it is more rapid in soils previously treated with the herbicide. High herbicide doses and repeated applications over a long period of are favorable for generating accelerated biodegradation. The accelerated biodegradation may reduce herbicide efficiency. On the other hand, it decreases the concern about the accumulation of herbicide in soil after repeated past applications (Bulcke et al., 2000).

Bogarets et al. (2000) studied diuron's microbial degradation and ecotoxicology to investigate its fate after application to soils. Quantitative biodegradation assays were executed with fungal strains, showing that diuron was degraded but not entirely mineralized. A series of tests were carried out to choose the most efficient fungal strain for diuron degradation. Among the fungal strains tested, only three were able to transform diuron to any extent (up to 50%) after 7 days of incubation: *B. bassiana*, *C. elegans*, and *M. isabellina*. No degradation occurred using fungal strains: *F. oxysporum* and *G. candidum*. Although *C. elegans* was the most efficient since there was no diuron remaining after seven days of incubation. Diuron degradation by the three fungal strains led to the formation of two metabolites obtained in different ratios according to the microorganism. For the three fungal strains, diuron degradation led to the formation of the demethylated products. The identified metabolites were synthesized in sufficient amounts to confirm their structures and determine their non-target toxicity using four biotests. According to the Microtox test, the metabolites N-(3,4-dichlorophenyl)-N-methylurea and N-3, 4-dichlorophenylurea presented a three times higher toxicity than that of diuron.

## **BIOTA**

*Plants.* Diuron is readily absorbed through the root system of plants and less readily through the leaves and stems. Diuron is translocated rapidly from roots to shoots via the xylem. Little to no diuron moves from the apex downward toward the base of a treated leaf via the phloem. Diuron symptoms of disease are foliar chlorosis concentrated around veins or sometimes interveinal followed by necrosis ((Hess and Warren, 2002).

In plants, diuron is metabolized via N-demethylation of the nitrogen atom and hydroxylation at position 2 of the benzene ring. Differential metabolism via N-demethylation may be the basis for diuron selectivity. For example, demethylation is catalyzed in cotton by an enzyme called N-demethylase. Diuron was metabolized to conjugates of monomethyl diuron in *Torilis arvenis* and to N-dealkylated derivatives in *Lolium rigidum* (Hess and Warren, 2002).

*Mammals.* Once diuron is ingested, it is excreted through feces and urine of test animals. In a study, cows fed very low doses of diuron in their diets had small amounts of residues in whole

milk. Cattle fed small amounts, accumulated low levels of diuron in fat, muscle, liver, and kidney. Little tissue storage under field conditions is anticipated (EXTOXNET, 1996).

In mammals, metabolism principally occurs through hydroxylation and dealkylation. The predominant metabolite in dogs is 3,4-dichlorophenyl urea. Breakdown of this compound is similar in animals, plants and soil. The first step is N-demethylation followed by ring cleavage (EXTOXNET, 1996).

*Fish and Aquatic Invertebrates.* Diuron is toxic to fish and aquatic invertebrates. The LC<sub>50</sub> (48 hr) values for diuron range from 4.3 to 42 mg/L in fish, and range from 1 to 2.5 mg/L for aquatic invertebrates. The LC<sub>50</sub> (96hr) is 3.5 mg/L for rainbow trout (EXTOXNET, 1996). Therefore diuron is moderately toxic to fish and to aquatic invertebrates.

## **SUMMARY**

A systemic phenylurea herbicide, diuron functions primarily by inhibiting the Hill reaction in photosynthesis. In California diuron is applied to both crop sites and noncrop sites, such as rights-of-way and industrial sites. Approximately 1,300,000 pounds of active ingredient were reported applied in California in 2002.

Sorption of diuron to soil is highly correlated with soil organic matter. However, relative to other pesticides diuron is generally considered both mobile and persistent. Diuron has been frequently detected in both California ground and surface water, and is regulated as a known ground water contaminant in California. Because diuron has a low Henry's law constant and a low vapor pressure, volatilization is considered insignificant.

Microbial degradation is considered to be the primary mechanism both in the dissipation from soil and in aquatic environments. Under aerobic conditions, diuron degrades by N-demethylation to the metabolites (1) N-(3,4-dichlorophenyl)-N-methylurea (DCPMU); (2) 3,4-dichlorophenylurea (DCPU); and (3) dichloroaniline (DCA). Under anaerobic soil conditions, a dechlorinated metabolite, N-(3-chlorophenyl)-N-methylurea (CPMU), is formed.

In plants, diuron is metabolized via N-demethylation of the nitrogen atom and hydroxylation at position 2 of the benzene ring. In mammals, metabolism principally occurs through hydroxylation and dealkylation. The predominant metabolite in dogs is 3,4-dichlorophenyl urea. Breakdown of the compound is similar in animals, plants and soil. Diuron is moderately to toxic to fish and aquatic invertebrates.

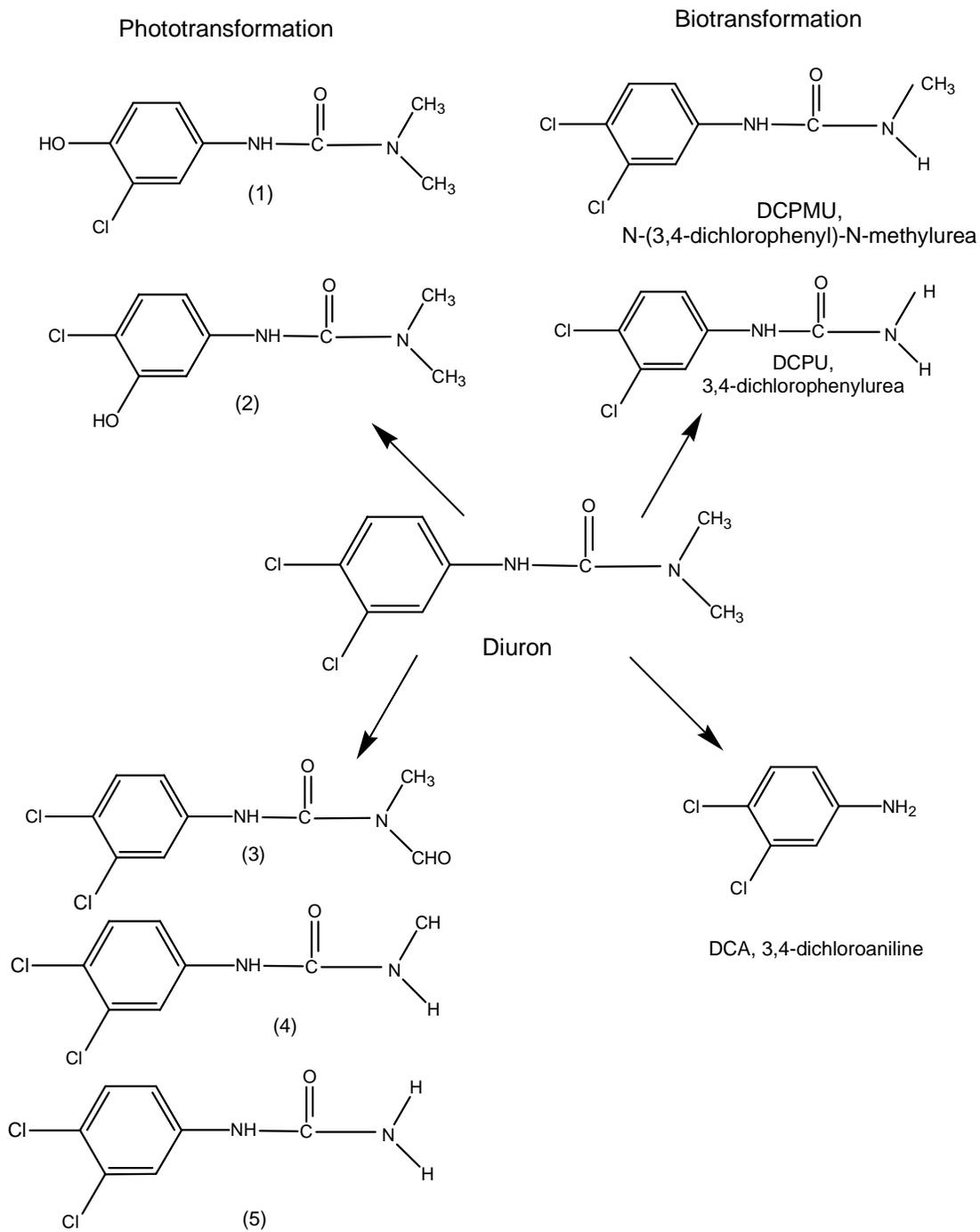
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**FIGURE 1. GENERAL SCHEME OF THE PHOTODEGRADATION AND BIOTRANSFORMATION**  
**Bonnemoy et al., 2001**



**FIGURE 2. ROUTES OF FORMATION OF PHOTOPRODUCTS OF DIURON**

**Boule et al., 1997**

