Leaching of Fluometuron and Diuron in a Vega Alta Soil[1]

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INTRODUCTION

The increased use of persistent pesticides has caused widespread alarm among those concerned with the quality of our environment. It is essential that we learn more about the movement and fate of pesticides in the total ecosystem. This is particularly important with respect to herbicides as they generally are considered to have greater mobility than insecticides in the environment. The movement of herbicides over or through the soil influences not only their effectiveness but also their potential as environmental contaminants. In a previous leaching study (7), it was found that 2 chloro-4-(ethylamino)-6-(isopropylamino)-s-triazine (Atrazine) had the greatest downward penetrativity, while 2,4-bis (isopropylamino)-6-(methylthio)-s-triazine (Prometryne) had the least. The mobility of 2-(ethylamino)-4-(isopropylamino)-6-(methylthio)-s-triazine (Ametryne) was found to be slightly greater than that of Prometryne. In the said study, oats (*Avena sativa* L.) was used as an indicator plant to assay the phytotoxic activity of herbicides at different depths. No attempt was made to study the herbicide movement in relation to possible environmental contamination. The present investigation represents an initial step designed to determine to what extent 3-(3,4-dichlorophenyl)-1,1-dimethylurea (Diuron) and 1,1-dimethyl-3-(α,α,α trifluoro-m-toyl) urea (Fluometuron) will leach in the soil from a series of lysimeters under simulated sugarcane field conditions.

REVIEW OF LITERATURE

Leaching of the substituted urea herbicides through the soil has been a subject of extensive investigation. Hill et al. (3) found that some leaching of 3-(p-chlorophenyl)-1,1-dimethylurea (Monuron) took place when an excessive high rate of herbicide and a large volume of water were applied

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to soil columns. However, under field conditions with an application rate of 1 to 2 pounds per acre, negligible amounts of Monuron and Diuron leaching by percolating water occurred below a depth of 4 inches. Upchurch and Pierce (9) demonstrated that Monuron could be displaced below 24 inches when applied at an abnormal high rate and leached with a large amount of water. Weldon and Timmon (11) studied the movement of Diuron (2 to 4 lb/A) in the soil and found that Diuron was held in the top few inches of the soil and was not carried into the soil 4 inches below the surface. Using the soil columns technique, Hilton and Yuen (4) failed to demonstrate the leaching of Diuron below a depth of 2 inches in a sugarcane topsoil treated with 20 pounds per acre. In the same study, Monuron, at rates of up to 10 pounds per acre, did not leach appreciably below 4 inches. Ivey and Andrew (5) reported that Diuron at 2.5 times the recommended rate stayed within the surface 6 inches in soil columns when percolated with 18 inches of simulated rain.

Appreciable leaching of Fluometuron was reported by Shahiedy and Andrew (8). They used the slotted column technique and found that Fluometuron penetrated to a maximum depth of 20 inches in a heavy Bowdre clay loam soil. Displacing Fluometuron and Diuron through saturated glass beads and soil, Davidson and Santelmann (1) found that the chemical was as mobile as the chloride ion at both high and low flow rates. However Diuron was not found to be quite as mobile. Most of the above leaching studies were conducted under laboratory and greenhouse conditions with little environmental contamination implications. Actually, the work of Edward et al. (2) on Methoxyclor and 2,4,5-T in lysimeter is the only type of study similar to ours. They found that the bulk of 2,4,5-T leaching loss took place within the first 4 months after application and that more than 50 percent of the loss occurred within 32 days. In contrast, the Methoxyclor concentration was generally lower for the first 3 months after application. A somewhat higher concentration prevailed from 4 to 10 months after application.

MATERIALS AND METHODS

A series of lysimeter units of the Ebermeyer type set up at the Station farm were used for conducting the present investigation. The individual lysimeter consists of flat funnels made of plastic material 16 inches in diameter and 2 inches deep with a 102 mm perforation in the center. The funnels were filled to the rim with quartz sand before setting them in the soil. A pit 3 feet deep, 3 feet wide and 10 feet long was dug in the selected site. Twelve of the stated funnels were buried in the soil on three sides of the pit at five different levels, e.g. 6, 12, 18, 24 and 36 inches from the surface. The arrangement and distribution of these funnels are shown in

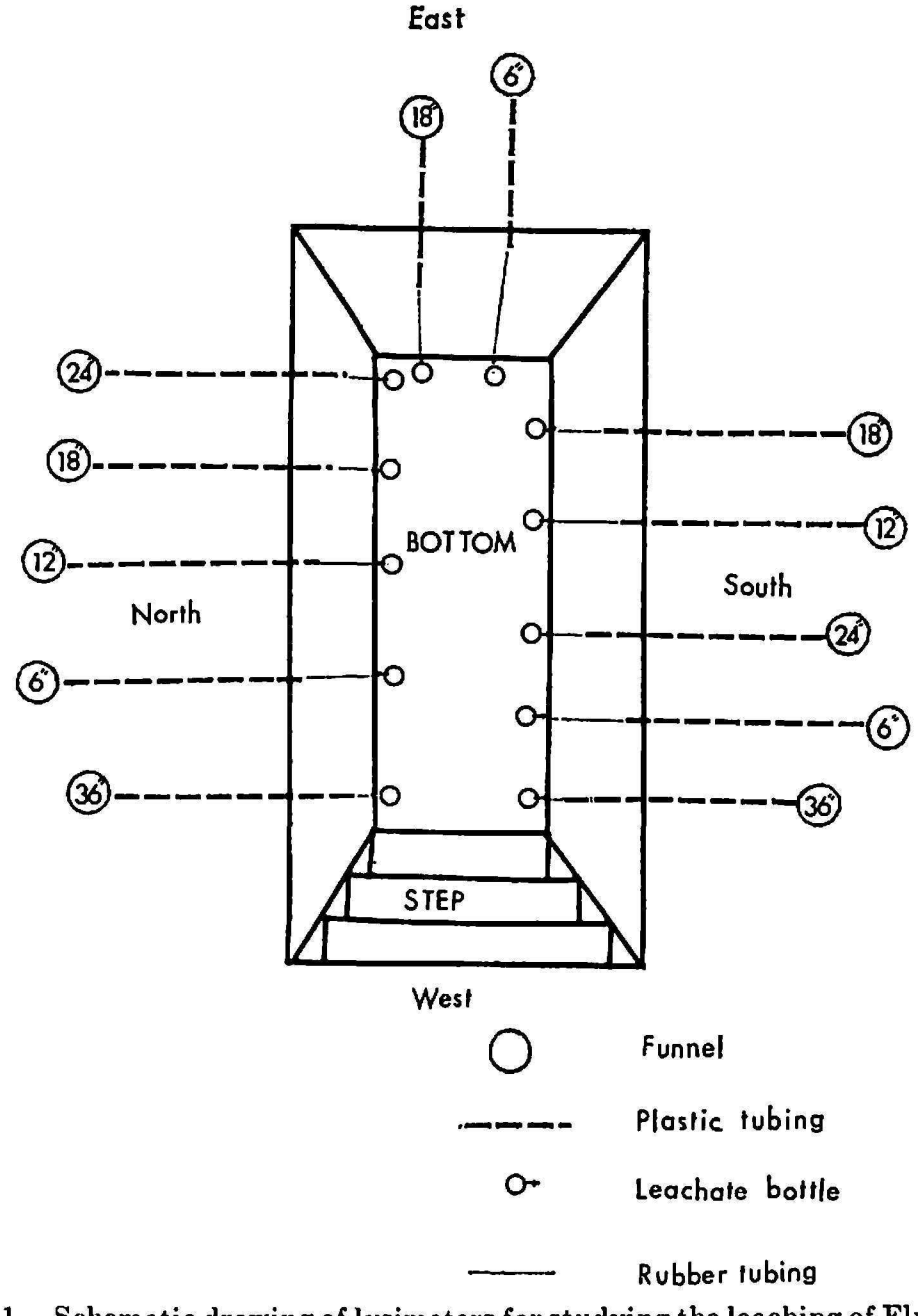

FIG. 1.—Schematic drawing of lysimeters for studying the leaching of Fluometuron and Diuron in a Vega Alta soil (Station Farm).

figure 1. As can be seen no two funnels occurred side by side at the same depth. The funnels were connected by means of plastic tubing to the respective leachate collecting bottles in the pit. The pit walls were lined with cinder-blocks and a shed roof placed over it to prevent rain from falling inside. The area adjoining the lysimeter installation was planted to sugarcane variety P.R. 980 (fig. 2).

The experiment was established on a Vega Alta clay soil. Vega Alta clay is a highly leached soil with friable brown or light brownish-gray heavy clay top soil. The bottom layers of this soil are reddish brown in color.

The average organic matter content, cation exchange capacity, clay content and pH for this soil are 2.02 percent, 15.0 meq per 100 g soil, 40.9 percent and 6.45, respectively.

The leachates were collected in 3-gallon bottles except those from the 24- and 34-inch depths, which due to the limited space available, were collected in 1-gallon bottles. The quantity of leachate collected was measured and recorded weekly. Two hundred ml samples were saved and stored in the refrigerator for the chemical determination of herbicides.

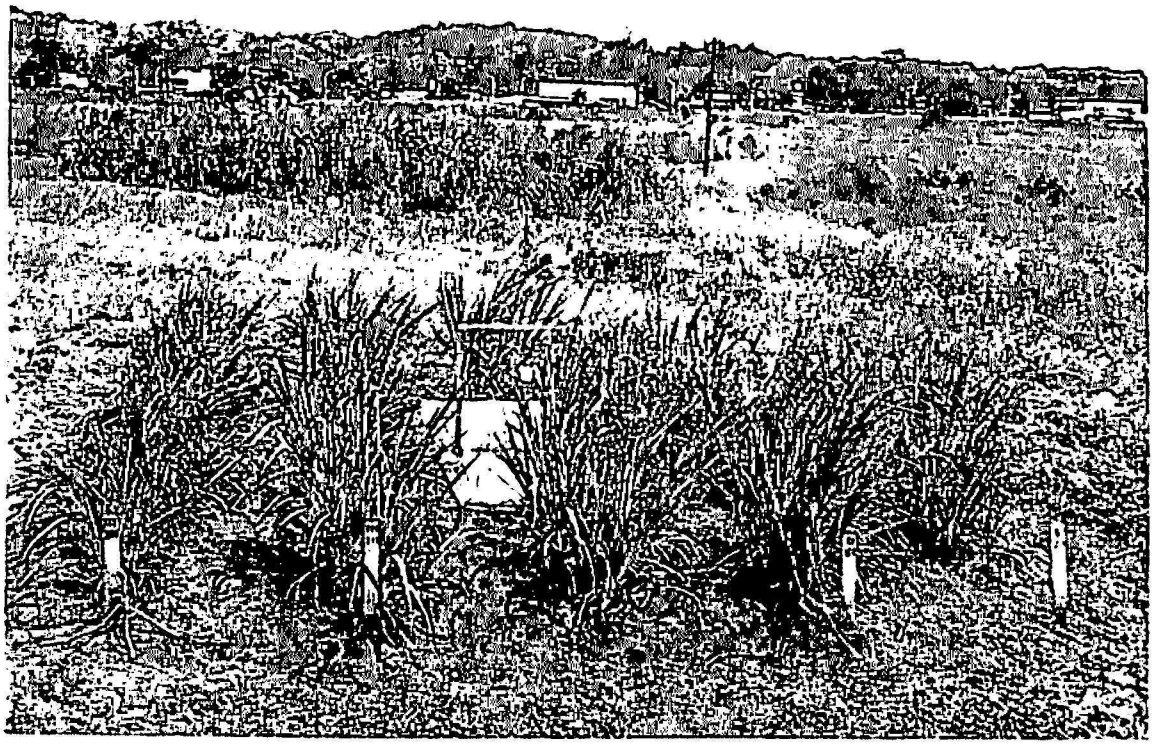

FIG. 2.—Appearance of the lysimeter used in the leaching study of herbicides.

Fluometuron at the rate of 4 pounds per acre was applied to plots established along the longer sides of the lysimeter in August 17, 1971. The narrower or eastern side was left untreated as a control or check. This experiment was carried on until the concentration of Fluometuron in the leachate fell below the detection limit of 0.05 p/m.

Afterwards, the sugarcane was harvested and newly planted for a Diuron experiment. Diuron in turn was applied at a per acre rate of 4 pounds in August 18, 1972. The procedure followed was that described for Fluometuron. Again, the experiment was discontinued when the Diuron concentration in the leachate fell below 0.03 p/m.

The analytical method described by Katz (2) for the chemical determina-

tion of Diuron and Fluometuron was followed. Diuron as well as Fluometuron were hydrolyzed under reflux conditions for 3 hours with 6*N* sulfuric acid. The resulting 3,4-dichloro aniline and 3-trifluoromethyl aniline were diazotized and reacted with N-(1-naphthyl) ethylene diamine hydrochloride. The magenta dye was extracted with n-butanol and sodium sulfate and the colored butanol layer read at 560 and 540 nm for 3,4-dichloro aniline and 3-trifluoromethyl aniline, respectively.

RESULTS AND DISCUSSION

The Diuron concentrations found in water samples from the lysimeter is graphically presented in figure 3. As it can be seen, the highest concentrations of Diuron prevailed during the first week of herbicide application. These concentrations, i.e., 0.59, 0.31, 0.21, 0.41 and 0.26 p/m were detected from 6-, 12-, 18-, 24- and 36-inch depth levels, respectively. The Diuron concentration at different depths decreased rapidly with the passage of time. All Diuron concentrations in water samples collected at different depths fell below 0.10 p/m after 4 weeks of application and tended to fluctuate somewhat as time advanced. The Diuron concentrations in the leachate at all depths were no longer detectable at the end of the 16th week. Similarly, the largest quantity of Diuron leaching loss was recorded during the first week of herbicide application (table 1). Approximately 80 percent of the total Diuron leaching loss occurred during this first week. Rainfall data show that a precipitation of 3.4 inches was registered in the first week after the herbicide application (table 2). The heavy rain might have contributed to the excessive leaching of this compound. The quantity of Diuron in the leachate again decreased drastically as time progressed. Finally, no detectable quantity of Diuron was found in the leachate from different depths at the end of 16th week. Although most leaching studies conducted under laboratory and greenhouse conditions (1,3,5,9,11) suggested the relative immobility of Diuron in the soil, it is apparent from our data that Diuron in low concentrations could penetrate to a maximum depth of 36 inches. Herbicides such as Monuron, Atrazine and Simazine, usually considered relatively immobile in soil, have been detected at depths of 1 to 2 feet in ground water (10).

The Fluometuron concentrations found in water samples from the lysimeter are illustrated in figure 3. As might be expected, the highest concentrations of Fluometuron were detected in water samples taken 1 week after the initial herbicide application. These concentrations were 0.70, 0.37, 0.33, 0.15 and 0.38 for 6-, 12-, 18-, 24- and 36-inch depth levels, respectively. A slower rate of Fluometuron concentration decline than that of Diuron was noted. Most of the Fluometuron concentration in water samples tended to fall below 0.10 p/m from the 13th week on. Fluometuron

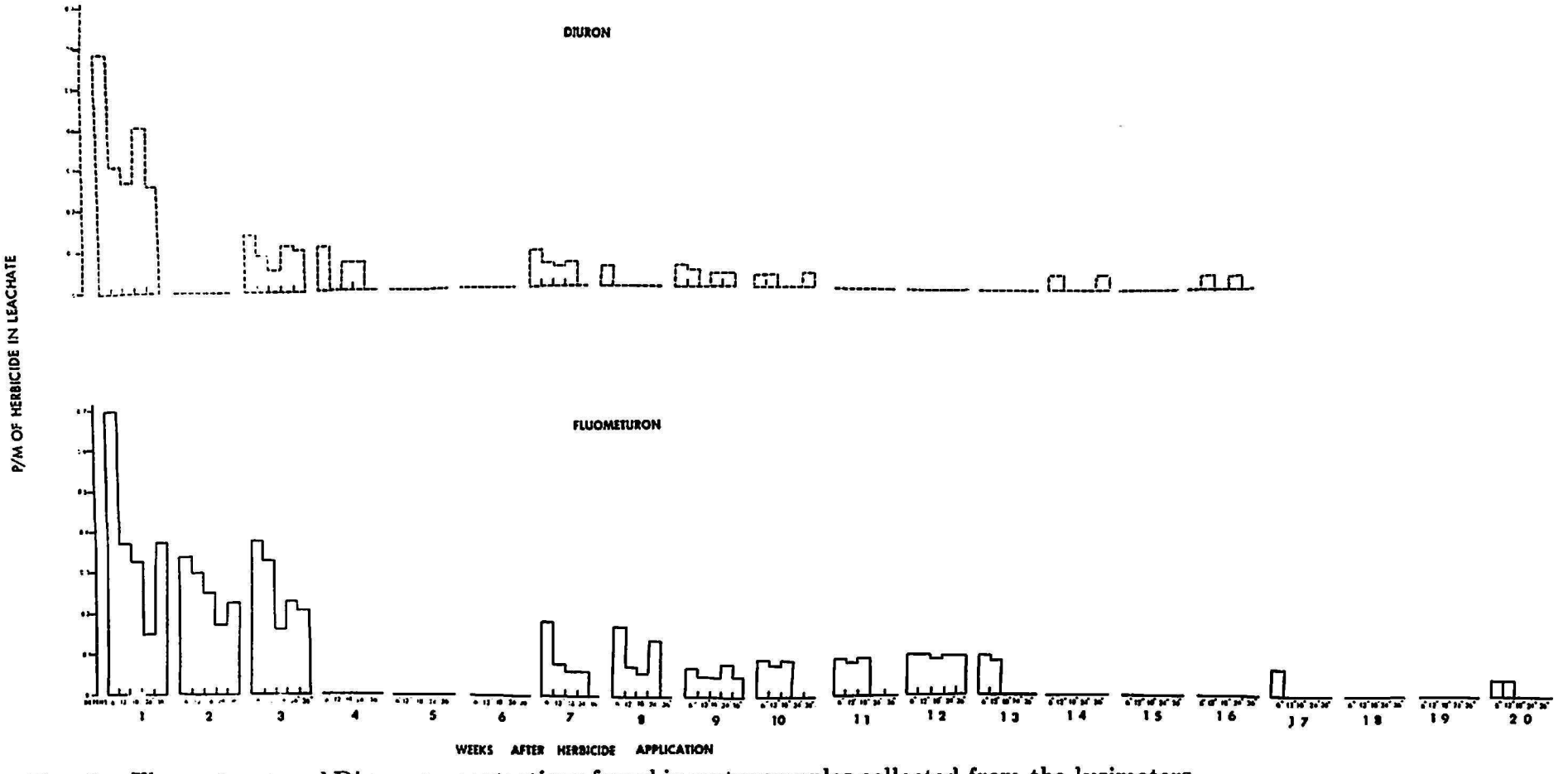

FIG. 3.—Fluometuron and Diuron concentrations found in water samples collected from the lysimeters.

concentration at all depths fell below the detection limit at the end of the 20th week after the herbicide was applied. The greatest leaching loss from the site of application of Fluometuron took place in the second week (table 3). It is worth noting that 3.8 inches of rainfall were recorded in this second week (table 2). The relationship between the quantity of herbicide leaching loss and the amount of rainfall is not fully known and has yet to be estab-

TABLE 1.—*Quantity of Diuron collected in leachate from the lysimeter*

Time in weeks after herbicide application	Quantity of Diuron at different depths (μg)					
	6 inches	12 inches	18 inches	24 inches	30 inches	Total
1	5,844.6[a]	2,846.8	4,600.8	3,176.0	666.6	17,134.8
2	—[b]	—	—	—	—	—
3	1,007.3	630.7	235.1	248.7	48.5	2,170.3
4	120.0	—	43.3	66.6	—	229.9
5	—	—	—	—	—	—
6	—	—	—	—	—	—
7	185.0	44.4	38.4	37.2	—	305.0
8	79.5	0	0	0	0	79.5
9	267.4	118.4	0	155.3	88.8	629.9
10	115.4	43.7	0	0	75.4	234.5
11	0	0	0	0	0	0
12	0	0	0	0	0	0
13	—	—	—	—	—	—
14	131.8	0	0	0	89.1	220.9
15	—	—	—	—	—	—
16	0	2.2	0	5.2	0	7.4
17	0	0	0	0	0	0
18	—	—	—	—	—	—
19	—	—	—	—	—	—
20	0	0	0	0	0	0
Total	7,751.0	3,686.2	4,917.6	3,689.0	968.4	21,012.2

[a] Each figure is the summation of quantity of Diuron from two collection pans.
[b] No water samples were collected during this period.

lished. The quantity of Fluometuron loss also tended to fluctuate somewhat with time. However, by the end of the 20th week, the quantity of Fluometuron in the water samples was beyond the detection limit. The relative ease with which Fluometuron leached from the soil in this study agrees with the findings of Ivey and Andrew (5) and Davidson and Santelmann (1).

On the basis of the above information on the concentration levels found in the water samples, it is evident that Diuron could pose a contamination problem only within the first 3 weeks of application. As for Fluometuron, the critical period could extend to within 12 weeks of application. However,

the concentration of a given herbicide in the water samples is a function of the total volume of water collected. Dilution factors could greatly change the concentration detected at different depth levels. Under test conditions, it appears that the quantity of herbicide lost from leaching is a better index of evaluation than concentration. Assuming that the downward leaching of Diuron and Fluometuron occurred with little or insignificant lateral movement, each one of the collection pans used in this study collected the herbicide applied directly above it. Thus, on the basis of a 4-pound per

TABLE 2.—*Inches of precipitation during the years 1971 and 1972*

Week	1971 for Fluometuron experiment	1972 for Diuron experiment
1	2.80	3.36
2	3.83	.81
3	2.16	2.47
4	Trace	.97
5	.29	.40
6	.28	.22
7	1.30	.72
8	1.93	1.03
9	1.95	1.62
10	1.37	.55
11	1.29	2.11
12	.50	.86
13	1.38	.59
14	.22	.96
15	.38	.84
16	1.17	1.89
17	1.16	3.75
18	1.91	.28
19	.56	.36
20	2.16	1.68
21	2.18	—
22	.56	—

acre herbicide application the 10 collecting pans could theoretically collect a total of 582,123 μg if there was a complete leaching of the material applied. However, the total amount of either Fluometuron or Diuron collected from the leachate during the course of the investigations was found to be 21,012 μg for the former and 32,153 μg for the latter herbicide. These only amounted to 3.6 and 5.5 percent of the total application for Diuron and Fluometuron, respectively. It seems that the expressed amounts do not pose a direct menace to ground water contamination. Moreover, the rapid decline in the detectable amounts of both herbicides in the collected water samples suggest that this contamination may be of a very short nature.

TABLE 3.—*Quantity of Fluometuron collected in leachate from the lysimeter*

Time in weeks after herbicide application	Quantity of Fluometuron at different depths (μg)					
	6 inches	12 inches	18 inches	24 inches	36 inches	Total
1	4,217.0[a]	2,278.4	239.8	404.8	727.8	7,867.0
2	4,544.0	2,360.1	3,779.0	1,419.2	694.6	12,796.9
3	2,988.7	1,643.4	798.6	1,646.4	425.8	7,502.9
4	—[b]	—	—	—	—	—
5	—	—	—	—	—	—
6	—	—	—	—	—	—
7	237.2	128.1	12.6	75.7	—	453.6
8	647.2	125.4	102.4	119.3	—	994.3
9	399.3	173.8	35.3	157.0	38.5	803.9
10	165.5	114.8	25.5	—	—	305.8
11	437.8	348.5	113.9	190.0	73.0	1,163.2
12	18.7	28.4	—	—	—	47.1
13	0	0	0	0	0	0
14	—	—	—	—	—	—
15	—	—	—	—	—	—
16	0	0	—	—	—	—
17	54.4	—	—	—	—	54.4
18	0	0	—	—	—	—
19	—	—	—	—	—	—
20	100.0	64.3	—	—	—	164.3
21	0	0	0	0	0	0
22	0	0	0	0	0	0
Total	13,809.8	7,265.2	5,107.1	4,011.6	1,959.7	32,153.4

[a] Each figure is the summation of quantity of Fluometuron from two collection pans.

[b] No water samples were collected during this period.

The accelerated rate of microbial activity under tropical conditions could further reduce the possible contamination hazard to a minimum.

SUMMARY

Leaching of Diuron and Fluometuron in field lysimeters was conducted during 1971 and 1972. Diuron and Fluometuron in low concentrations were found to leach to a maximum depth of 36 inches. The highest concentrations of both herbicides were detected in water samples taken 1 week after herbicide application. The greatest quantity of Diuron lost from leaching occurred during the first week of its application; Fluometuron in the second week. The leaching loss of Diuron was no longer detectable 16 weeks after application; Fluometuron, 20 weeks. The leaching loss of Diuron and Fluometuron was estimated to be 3.6 and 5.5 percent, respectively, of the total application.

RESUMEN

En 1971 y 1972 se realizó un estudio para determinar la lixiviación de los herbicidas Diuron y Fluometuron en lisímetros de campo. El Diuron y el Fluometuron penetraron, a bajas concentraciones, a una profundidad máxima de 36 pulgadas en el suelo. La concentración más alta de ambos herbicidas en el agua apareció en las muestras que se tomaron una semana después de su aplicación. La mayor pérdida de Diuron por lixiviación tuvo lugar durante la primera semana siguiente a la aplicación y la de Fluometuron durante la segunda semana. La pérdida de Diuron por lixiviación dejó de detectarse 16 semanas después de su aplicación y la de Fluometuron después de 20 semanas. La cantidad de Diuron y Fluometuron perdida por lixiviación se estimó en 3.6 y 5.5 por ciento, respectivamente, del total aplicado.

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