

Effect of Soil Characteristics on Adsorption and Mobility of (¹⁴C)Diazinon[†]

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The adsorption and mobility of diazinon (*O,O*-diethyl *O*-2-isopropyl-6-methylpyrimidin-4-yl phosphorothioate) in 25 soils of different physicochemical properties were studied. The adsorption isotherms were found to conform to the Freundlich adsorption equation. Freundlich's constant, K , and the distribution coefficient, K_d , were found to be highly significantly correlated ($p < 0.001$) with the organic matter (OM) content when all soils or only those with an OM content above 2% were considered. There was also a significant correlation ($p < 0.01$) of K and K_d with the silt plus clay content of soils with an OM content below 2%. On the basis of the R_f values obtained by TLC, the pesticide was found to be slightly mobile in 80% and immobile in 20% of the soils studied. A study of the pesticide mobility in soil columns under saturated flow conditions showed it to be leached from light soils with low OM contents.

Keywords: Diazinon; soil; adsorption; mobility; TLC; soil columns

INTRODUCTION

Adsorption and mobility of pesticides in soils are important processes involved in the environmental behavior of these compounds (Hamaker and Thompson, 1972; Khan, 1980; Koskinen and Harper, 1990). This, together with the increasingly frequent occurrence of pesticides in the surface and ground waters of different countries (Hallberg, 1989; Leistra and Boesten, 1989; Legrand et al., 1991), has drawn the attention of environmental scientists to such processes.

Studies on the adsorption and mobility of pesticides in soil have so far focused on herbicides in preference over insecticides. With regard to the broad group of organophosphorus insecticides, adsorption of some of these compounds by soils has been studied by several authors (MacNamara and Toth, 1970; Felsot and Dahm, 1979; Sharom et al., 1980; Reddy and Cambrell, 1987; Sánchez-Camazano and Sánchez-Martín, 1984, 1990), as has been their mobility in soils (Khan and Khan, 1986; Kumari et al., 1991).

Diazinon (*O,O*-diethyl *O*-2-isopropyl-6-methylpyrimidin-4-yl phosphorothioate) is a nonsystemic organophosphorus insecticide widely used for controlling the impact of insects on horticultural, beet, cotton plant, and rice crops. Several authors have reported on its presence in underground waters (Hallberg, 1989), its adsorption by individual components of soils, clay, and humic acid (Saint-Fort and Visser, 1988; Dios et al., 1984), the formation of interlamellar compounds with cation-saturated smectite (Dios et al., 1985), its persistence in soils relative to other pesticides (Sattar, 1990), and its mobility (Somasundaram et al., 1991) and adsorption (Leistra et al., 1984) in a limited number of soils. However, the influence of soil parameters on the adsorption and mobility of this pesticide had not yet been statistically studied on a reasonably large number of soils.

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Diazinon is widely used for treating cotton, beet, and rice crops in the Marismas del Guadalquivir, a lowland area in west-south Spain. The intensive, highly productive farming of the region calls for large amounts of pesticides. Determining the mobility of pesticides in the soils of this region is of special interest because it is close to the Doñana National Park, one of the finest wetlands in Europe. The use of pesticides in the region poses serious pollution hazards to the park waters, where some authors have shown the presence of several pesticides (Albaiges et al., 1985; Baluja et al., 1985).

In this work, the adsorption and mobility of diazinon in 25 soils and the influence of soil properties on both processes were studied. Twenty of the soils were from the Marismas del Guadalquivir, while the other 5 were natural, uncultured soils from the province of Salamanca (west-central Spain) and contained over 3% of organic matter (OM).

MATERIALS AND METHODS

Materials. ¹⁴C-Labeled diazinon of specific activity 185 MBq mg⁻¹ and 97% purity was obtained from International Isotope (Munich, Germany). Unlabeled diazinon of technical purity (98%) was supplied by Riedel de Haen (Hannover, Germany). Diazinon is a liquid with water solubility of 40 mg L⁻¹; the compound is also soluble in most organic solvents (Worthing and Walker, 1987).

Table 1 shows the types of soils used and some selected characteristics. Soils 1-20 were from the Marismas del Guadalquivir, while soils 21-25 were from the province of Salamanca. The latter, with OM contents above 3%, were included owing to the low OM contents (0.15-2.6%) of the Marismas soils and the high significance of this parameter to pesticide adsorption and mobility in soils. Samples were air-dried and sieved through 2-mm mesh. Their particle size distribution was determined by using the pipet method (Day, 1965). Organic carbon was determined according to the Walkley-Black modified method (Jackson, 1958), results being multiplied by 1.72 for conversion into OM contents. Soil pH values were measured in slurries made in a 1:1 soil/water ratio. Clay minerals were identified by the X-ray diffraction technique.

Adsorption Isotherms. The diazinon adsorption isotherms for the different soils were obtained by treating duplicate samples of 1 g of soil with 10 mL of an aqueous solution of the pesticide at a concentration of 5, 10, 15, 20, or

Table 1. Selected Characteristics of the Soils

soil	soil texture	soil type	pH	OM (%)	sand (%)	silt (%)	clay (%)	clay mineralogy ^a
Marismas								
1	silty clay	Typic Salorthid	7.7	0.82	4.4	53.0	42.6	I, S, K
2	clayed	Typic Salorthid	7.8	1.10	4.2	27.1	68.7	I, S, K
3	clayed	Typic Xerofluvent	7.6	1.55	11.2	26.7	62.1	I, S, K
4	clayed	Typic Xerofluvent	7.7	0.57	25.7	32.5	41.8	I, S, K
5	sandy clay loam	Typic Xerofluvent	7.8	2.16	26.3	21.9	51.8	I, S, K
6	sandy clay loam	Typic Xerofluvent	7.9	0.55	53.1	11.0	35.9	I, S, K
7	sandy clay loam	Typic Xerofluvent	7.9	0.47	70.9	5.3	23.8	I, S, K
8	sandy clay loam	Typic Rhodoxeralf	7.7	0.93	73.0	3.0	24.0	I, K, S
9	sandy clay	Typic Rhodoxeralf	7.4	0.57	53.1	5.9	41.0	I, S, K
10	sandy clay loam	Dystric Xerochrept	4.6	0.15	65.4	5.6	29.0	I, K
11	sandy loam	Dystric Xerochrept	5.8	1.30	78.2	6.6	15.2	K, I
12	sandy loam	Dystric Xerochrept	4.6	0.22	75.8	5.7	18.5	I, K
13	sandy loam	Typic Haploxeralf	7.9	0.72	74.9	8.5	16.6	I, K, S
14	clayed	Entic Chromoxerert	7.4	2.01	33.1	14.3	52.6	I, K, S
15	clayed	Entic Chromoxerert	7.4	2.60	13.5	23.9	62.6	I, S, K
16	sandy clay loam	Entic Pelloxerert	7.9	1.89	59.6	12.2	28.2	I, K, S
17	sandy clay loam	Typic Fragixeralf	7.2	1.52	66.6	6.9	26.5	I, S, K
18	sandy clay loam	Typic Fragixeralf	7.9	0.52	65.9	7.5	26.6	I, S, K
19	sandy	Xeric Torripsamment	5.2	0.69	88.8	2.7	8.5	S, I, K
20	sandy loam	Typic Rhodoxeralf	7.5	0.77	66.4	15.5	18.1	I, K, S
Salamanca								
21	sandy loam	Typic Xerumbrept	4.7	10.20	64.3	21.1	14.6	I, K
22	sandy loam	Typic Xerumbrept	5.0	8.90	67.4	18.7	13.9	I, K, V
23	loamy sand	Typic Xerumbrept	5.3	5.95	75.4	13.0	11.6	I, K, V
24	loamy sand	Typic Xerumbrept	5.1	4.66	73.9	11.9	14.2	I, K, V
25	sandy loam	Dystric Xerorthent	5.6	3.44	54.2	34.0	11.8	I, K, V

^a In order of abundance: I, illite; S, smectite; K, kaolinite; V, vermiculite.

25 $\mu\text{g mL}^{-1}$ and an activity of 41 Bq mL^{-1} . The suspensions were kept at 20 °C for 24 h in a thermostated chamber with intermittent agitation (2 h every 6 h). Preliminary experiments showed contact for 24 h to be long enough for equilibrium to be reached. Subsequently, the suspensions were centrifuged at 5045g for 30 min. To determine diazinon concentration, a 1.0-mL aliquot of supernatant was then withdrawn from each tube and added to 4.5 mL of scintillation liquid, and its activity was measured in disintegrations per minute (dpm) on a Beckman LS 1800 liquid scintillation counter. The dpm recorded for the supernatant aliquot was related to the dpm obtained for the aliquots of the respective standards of diazinon solutions, and the diazinon equilibrium concentration was determined. The amount of pesticide adsorbed was considered to be difference between that initially present in solution and that remaining after equilibration with the soil. The least significant difference (lsd) test was used to determine significant differences among K values and among K_d values of different soils at the 5% significance level.

Soil Thin-Layer Chromatography (TLC). Soil plates for TLC were prepared by grinding the soil samples in a mortar, followed by sieving through 160- μm mesh. Soil (7.5 g) and distilled water (15 g) were slurried and spread as a 0.5-mm-thick layer over 20 \times 5 cm^2 glass plates with the aid of a Desaga TLC spreading device. Five plates were prepared for each soil, of which the first and fifth of them were not used. The selected plates were dried in a chamber at room temperature and subsequently stored in a desiccating chamber at a relative humidity of 70%. The plates were marked with two horizontal lines at distances of 2 and 12 cm, respectively, from the base. A 3- μL droplet of (¹⁴C)diazinon (550 Bq) was spotted on the baseline of the three plates with the aid of a micropipet. The plates were placed in closed individual glass chromatographic chambers that were 22 cm long and 5 cm wide. After distilled water migrated to a distance of 10 cm from the baseline, the plates were allowed to dry at room temperature. Movement of (¹⁴C)diazinon was detected by using a Berthold TLC Tracemaster 20 linear detector. The mobility factor, R_f , is given by $R_f = R_i/10$, where R_i is the frontal distance traveled by the insecticide. R_f values were subjected to analysis of variance at the 5% confidence level.

Soil Column Leaching. Leaching columns were constructed from poly(vinyl chloride) (PVC) pipe and were 40 cm long \times 5 cm id. They were prepared according to the Weber (1986) method. Each column was packed with 600 g of dry

soil and saturated by putting them into a tank and increasing the water volume in the tank every 24 h for 1 week until it topped the columns. Then, each column was allowed to drain free for 24 h. Five milliliters of a 200 $\mu\text{g mL}^{-1}$ ethanol solution of diazinon (5 kg ha^{-1}) of 36 kBq mL^{-1} specific activity was added to the top of each column. The columns were then washed with 200 mL (10.2 cm) of water on a daily basis for 12 days. The column leachates were monitored daily for the insecticide content by using 1 mL of sample for measuring the activity (dpm). After draining for some time, the columns were cut breadthwise at 5-cm intervals. The soil contained in each segment was turned over and weighed. Then, five samples of 1 g each were taken from every segment. Two such samples were used to determine the moisture content of the soil from the weight loss measured on calcination at 80 °C for 12 h. The other three samples were employed to determine the ¹⁴C content in a Harvey OX-500 biological oxidizer. The activity of the radioactive solutions containing ¹⁴CO₂ was determined with a Beckman LS 1800 scintillation counter by measuring the dpm for each sample. The experiment was performed in duplicate, and a pesticide-untreated column for each soil was subjected to the whole process in parallel to determine its natural radioactivity for reference.

RESULTS AND DISCUSSION

The adsorption isotherms of diazinon by soils show different forms according to soil characteristics (Figure 1). Those for soils with an OM content not greater than 0.77% were S-type (initial convex curvature) according to the classification of Giles et al. (1960); *i.e.*, they reflected a low adsorbent-adsorbate affinity at low concentrations. The samples with OM contents above 0.77% provided L-type isotherms (initial concave curvature), which are typical of a high adsorbent-adsorbate affinity. The isotherms obtained for diazinon adsorbed in humic acid of various molecular weights are also L-type (Saint-Fort and Visser, 1988). Even though the initial concave and convex curvatures of the isotherms obtained in this work were smooth (all of them approached type C), their shapes showed that the affinity of the soils for diazinon was related to their organic matter content.

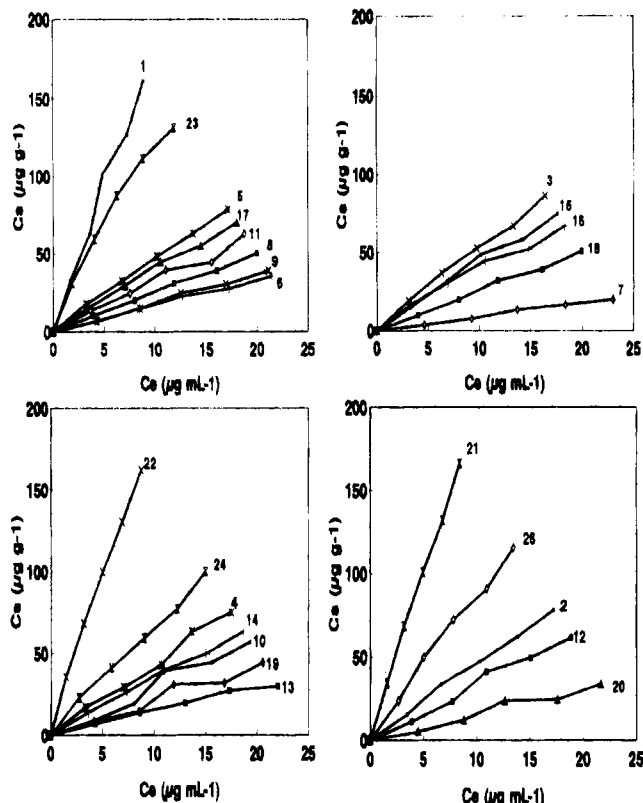


Figure 1. Adsorption isotherms of diazinon on soils.

The isotherms obtained generally conformed to the Freundlich adsorption equation, with a correlation coefficient $r > 0.97$. This equation can be expressed in linear form as

$$\log C_s = \log K + 1/n \log C_e$$

where C_s ($\mu\text{g g}^{-1}$) is the amount of adsorbed pesticide, C_e is the equilibrium concentration of dissolved pesticide ($\mu\text{g mL}^{-1}$), and K and n are two constants characteristic of the pesticide adsorption capacity. In fact, K is the amount of pesticide adsorbed at an equilibrium concentration of $1 \mu\text{g mL}^{-1}$, so it represents adsorption at low concentrations. On the other hand, n reflects to what extent the pesticide adsorption is dependent on the concentration. The K and n values obtained from the isotherms, as well as K_d and $\log K_{om}$ values, are given in Table 2. The distribution coefficient, K_d , represents adsorption at equilibrium concentrations higher than K and is defined as the ratio between the pesticide concentration in the soil and that in the equilibrium solution at a given equilibrium concentration (K_d was calculated for $C_e = 8 \mu\text{g mL}^{-1}$). Finally, K_{om} is the K value normalized to 100% organic matter [$K_{om} = 100 \times (K/\%OM)$].

As can be seen in Table 2, the largest K values corresponded to soils 21–25, with OM contents above 3%. Among the Marismas soils (1–20), the largest K values corresponded to soils 1 and 3, which, despite their low and medium OM content, respectively, contained abundant silt plus clay. K and K_d values were very similar since n was essentially unity in all cases. Values of n greater than 1 corresponded to samples that exhibited L-type isotherms, whereas n values below unity corresponded to soils giving rise to S-type isotherms.

The K values for diazinon obtained in this work were smaller than those reported for other organophosphorus

Table 2. Constants and Correlation Coefficients of the Freundlich Adsorption Equations (K , n , and r), Distribution Coefficients (K_d), $\log K_{om}$, and R_f Values

soil	K	n	r	K_d^a	$\log K_{om}$	R_f
1	20.04	1.05	0.999	18.14	3.39	0.18
2	4.83	1.02	0.998	4.64	2.64	0.19
3	7.15	1.13	0.999	5.56	2.66	0.22
4	5.35	1.06	0.998	4.45	2.97	0.17
5	6.25	1.13	0.997	4.90	2.46	0.17
6	1.58	0.97	0.998	1.66	2.46	0.23
7	0.70	0.92	0.999	0.84	2.17	0.23
8	2.74	1.03	0.997	2.57	2.47	0.23
9	1.46	0.92	0.996	1.75	2.41	0.23
10	1.58	0.80	0.987	2.57	3.02	0.12
11	4.14	1.10	0.999	3.41	2.50	0.14
12	2.58	0.91	0.997	3.16	3.07	0.17
13	1.79	1.07	0.998	1.54	2.40	0.27
14	4.20	1.08	0.999	3.58	2.32	0.13
15	5.11	1.06	0.998	4.49	2.29	0.12
16	6.57	1.26	0.999	4.25	2.54	0.14
17	5.15	1.11	0.998	4.19	2.53	0.19
18	2.45	0.99	0.997	2.52	2.67	0.17
19	1.75	0.94	0.979	2.00	2.40	0.10
20	0.93	0.85	0.989	1.36	2.08	0.15
21	22.73	1.07	0.999	19.72	2.35	0.07
22	25.73	1.18	0.999	18.48	2.46	0.07
23	19.76	1.27	0.998	12.64	2.52	0.09
24	9.42	1.17	0.999	6.88	2.31	0.08
25	9.83	1.05	0.999	8.90	2.46	0.09
lsd _{0.05} =	2.01			0.80		0.05

^a $C_e = 8 \mu\text{g mL}^{-1}$.

Table 3. Simple Correlation Coefficients (r) between Freundlich Constant (K) and Distribution Coefficient (K_d) and Soil Characteristics

soil	no. of soils	constant	pH	OM	clay	silt + clay
all soils	25	K	-0.41 ^a	0.84 ^c	-0.24	0.14
		K_d	-0.40 ^a	0.80 ^c	-0.20	0.20
OM >2%	8	K	-0.80 ^a	0.94 ^c	-0.71 ^a	-0.69 ^a
		K_d	-0.81 ^a	0.97 ^c	-0.69 ^a	-0.61
OM <2%	17	K	0.21	0.33	0.34	0.68 ^b
		K_d	0.13	0.20	0.33	0.68 ^b
Marismas	20	K	0.22	0.28	0.32	0.64 ^b
		K_d	0.14	0.17	0.30	0.62 ^b

^a Significant at 0.05–0.01 level. ^b Significant at 0.01–0.001 level. ^c Significant at <0.001 level.

insecticides including chlorpyrifos, phorate, and terbufos (Felsot and Dahm, 1979; Sharom et al., 1980) but greater than those reported for mevinphos (Sharom et al., 1980; Sánchez-Camazano and Sánchez-Martín, 1990). With regard to parathion, some of its reported K values exceed that for diazinon (Felsot and Dahm, 1979; Sharom et al., 1980), while other are smaller (Saltzman and Yaron, 1972; Sánchez-Martín and Sánchez-Camazano, 1991a).

The influence of soil components on diazinon adsorption was determined by using a statistical approach. The single correlations between K and K_d and the different chemical and texture properties of the soils were determined by least-squares linear regression analysis (Table 3). First, all soils were considered together. Then, two soil groups were considered, viz. soils with an OM content above 2% and soils with an OM content below 2%.

When all of the soils were considered, a highly significant correlation ($p < 0.001$) was found between both K and K_d and the OM content; however, no correlation was observed between K and K_d and the clay or silt plus clay content. This indicates that the OM

content is the most significant parameter in the adsorption of diazinon in soils with widely variable OM, silt, and clay contents. On the basis of the determination coefficient, r^2 , the OM content accounted for 70.5% of the variance of adsorption.

The correlation coefficient was also quite high and highly significant ($p < 0.001$) when only the soils with an OM content above 2% were considered. The determination coefficient for correlation between K_d and the OM content accounted for 94% of the variance of adsorption. Therefore, diazinon adsorption in soils with a high OM content depends almost solely on such a content.

No correlation between K or K_d and the OM content was found for the group of Marismas soils and for the group with OM content below 2%. There was, however, a significant correlation ($p < 0.01$) of K and K_d with the silt plus clay content of the soils. This indicates that when the OM content is low, the significance of other soil properties on diazinon adsorption is revealed.

The results for each soil group were also subjected to multiple linear regression analysis by combining two or more variables. The correlation coefficients (R^2) obtained provided no further explanation for the adsorption variability relative to the single correlation coefficients (r^2), though.

The above results suggest that no generalization can be made on the prominent role of organic matter in diazinon adsorption by soils. Previous studies also revealed significant correlations between adsorption and organic matter for widely variable OM contents and scarcely significant or no correlations for OM contents below 2% (Calvet et al., 1980; Sánchez-Camazano and Sánchez-Martín, 1984; Sánchez-Martín and Sánchez-Camazano, 1991b).

K or K_d was also negatively correlated with the soil pH. This was a result of the significant, though negative, correlation between the OM content and pH found by considering all of the soils or only those with an OM content above 2% [$r = -0.52$ ($p < 0.01$) and $r = -0.83$ ($p < 0.01$), respectively].

Log K_{om} values, which are very frequently used as a measure of the pesticide adsorption capacity of soils, ranged from 2.08 to 3.39. Consistent with the correlation between K and the OM content, the K_{om} range was quite narrow. The largest values (close to or greater than 3), which were also the most deviant from the more frequent value (2.46), corresponded to soils 1 and 4, which had low OM contents and high silt plus clay contents, and to soils 10 and 12, with the lowest OM contents of all. The log K_{om} values for the soils with OM contents above 2% were very similar (2.31–2.52), again indicating pesticide sorption occurrence largely on organic matter. The average log K_{om} values obtained by other authors for three soils (Briggs, 1981; Green and Karickhoff, 1990) were 2.50 and 2.52, respectively. These values lie within the range of log K_{om} values found in this work. The log K_{om} value calculated from log K_{ow} for diazinon [3.11 according to Briggs (1981)], by using the Briggs (1981) equation was 2.26, which also lies in the log K_{om} range obtained in this work.

Table 2 lists the R_f values obtained from soil TLC (averages of three determinations) used to study diazinon mobility. If all of the soils are considered, then R_f varied between 0.07 and 0.27 (mean = 0.16). For the soils with OM contents above 3%, R_f ranged from 0.07 to 0.09 (mean = 0.08). According to the classification of Helling and Turner (1968), diazinon can be considered

Table 4. Simple Correlation Coefficients (r) between R_f and Soil Characteristics

parameter	all soils	OM > 2%	OM < 2%	Marismas
K	-0.57 ^b	-0.77 ^a	-0.06	-0.07
K_d	-0.54 ^b	-0.76 ^a	-0.08	-0.08
pH	0.72 ^c	0.89 ^b	0.61 ^b	0.51 ^a
OM	-0.68 ^c	-0.83 ^b	-0.02	-0.28
clay	0.36	0.65 ^a	0.32	0.03
clay + silt	0.15	0.67 ^b	0.20	-0.001

^a Significant at 0.05–0.01 level. ^b Significant at 0.01–0.001 level. ^c Significant at <0.001 level.

slightly mobile in the soils with OM content below 3% and immobile in the soils with OM content above 3%.

Table 4 gives the single correlations obtained between R_f and the soil properties, K and K_d . If all of the soils are considered, there is a highly significant ($p < 0.001$) negative correlation between R_f and the OM content, as well as a very significant ($p < 0.01$) negative correlation of R_f with K and K_d . These correlations were also observed in the group of soils with OM contents above 2% but not in the soils with OM < 2% or those from the Marismas. In every case, however, there was a significant correlation between R_f and pH which can be ascribed to the significant correlation between the OM content and pH in both cases, as stated above in the description of the adsorption process. R_f values (0.10–0.24) similar to those obtained in this work were previously reported by Somasundaram et al. (1991), who studied the pesticide mobility in six soils with OM contents between 0.7 and 6.1%, *i.e.*, similar to those of our soils.

Under the conditions of study by TLC of diazinon mobility (saturated flow, a single leaching), diazinon was found to be slightly mobile in 80% of the soils and immobile in 20% of them. Because the TLC plates could not be subjected to successive leaching without causing the soil film to crumble, we believed it of interest to study mobility in packed columns subjected to successive leaching, *i.e.*, under the saturated–nonsaturated flow conditions used by Weber and Whitacre (1982). For this purpose, we chose two light soils containing little organic matter (soils 19 and 20) and hence having low K values; according to the TLC study, diazinon would be slightly mobile in them. Consequently, the two soils should lend themselves readily to leaching of the pesticide.

Figure 2 shows the variation of the diazinon concentrations and cumulative amounts found in the column leachates with the water volume leached. The cumulative amount was greater for soil 19 (38.5%) than for soil 20 (30.4%). The peak concentrations of diazinon were 4.7% for soil 19 and 7.0 for soil 20. The lag of the peak for soil 20 relative to soil 19 was only 165 mL.

The amount of diazinon retained in the column was smaller for soil 19 (29.3%) than it was for soil 20 (36.7%). The pesticide distribution in the different column segments is graphically depicted in Figure 3. The first segment of soil 20 contained a higher diazinon concentration than soil 19. The pesticide distributions in the other column segments were very similar for the two soils. Even though both were light soils, the differences in their texture reflected the diazinon amounts found both in the leachates and in the soils retained in the columns.

The results obtained with the columns show the importance of the water flow to pesticide mobility in light soils containing little organic matter.

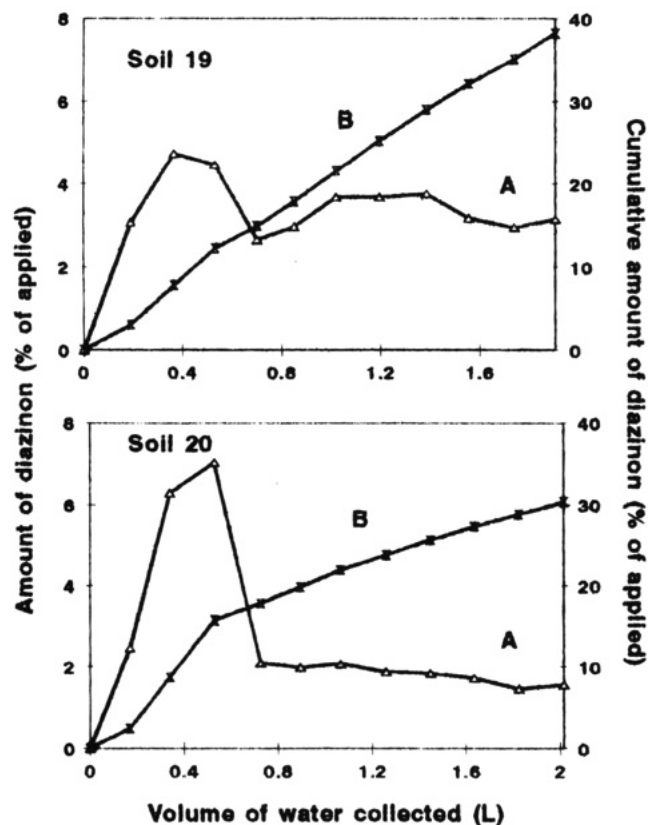


Figure 2. Measurement of diazinon in leachates as function of water collected: concentrations (A) and cumulative amounts (B).

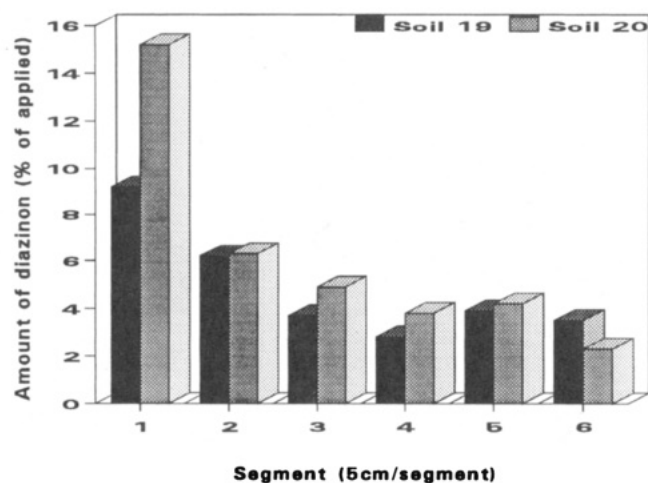


Figure 3. Distribution of diazinon in the soil columns.

CONCLUSIONS

The results obtained in this work show the OM content to be the most influential parameter on diazinon adsorption by soils containing over 2% OM. On the other hand, adsorption of the pesticide in soils with OM content below 2% is influenced by the silt plus clay content of the soil. On the basis of the mobility results obtained by TLC (*i.e.*, by using saturated flow conditions and a single washing), diazinon is slightly mobile in soils with a low or medium OM content and immobile in those with a high OM content. Leaching the packed columns under saturated–nonsaturated flow conditions, which involved several washings under saturated flow (drastic) conditions, showed the pesticide to be quite mobile in soils of light texture containing little organic matter. These results reflect the influence of the water

flow on diazinon mobility; under nonsaturated flow conditions, which are more similar to natural conditions, the pesticide should not be easily leached from the studied soils to ground water.

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