

# Mobility and Persistence of Isazofos in Granular and Microencapsulated Formulations in Two Soils, using Field Lysimeters

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(Revised manuscript received 19 May 1992; accepted 14 September 1992)

**Abstract:** Field lysimeters were used to assess the mobility and persistence of microencapsulated and granular formulations of the soil insecticide, isazofos, in Plainfield sand, and the microencapsulated formulation in Vittoria loam soil, using two moisture regimes, rainfall and supplementary watering. Mobility and persistence comparisons were made with an earlier lysimeter study which used emulsifiable concentrate (EC) and granular formulations of isazofos in Plainfield sand. Isazofos mobility in Plainfield sand increased in the following order for the tested formulations: microencapsulated < granular < EC. Atrazine, which was applied as a suspension concentrate to all lysimeters as an internal reference, appeared to exhibit retarded disappearance rates during initial stages of the study when in the presence of granular isazofos in the rainfall treatment. The degradation of isazofos was faster in Vittoria loam than in Plainfield sand for the microencapsulated formulation in the field lysimeters (only formulation tested), and for all three formulations in a laboratory study.

## 1 INTRODUCTION

The type of formulation can strongly influence mobility and, to a certain degree, the persistence of pesticides in soil. In an age when it is very difficult and costly to register new pesticides, and when environmental pressures are curtailing current use patterns, formulation technology may be a partial solution for making selected pesticides more useful and environmentally safe. In an earlier field lysimeter study,<sup>1</sup> we examined the mobility and persistence characteristics of emulsifiable concentrate (EC) and granular formulations of isazofos (O-5-chloro-l-isopropyl-1 H-1,2,4-triazol-3-yl 0,0-diethyl phosphorothioate) in Plainfield sand. In this study we test a new microencapsulated formulation of isazofos in both Plainfield sand and Vittoria loam soils, using a set of lysimeters treated with the granular isazofos formulation as a link between the studies.

## 2 MATERIALS AND METHODS

### 2.1 Compounds and application rates

Atrazine as a commercial 280 g litre<sup>-1</sup> suspension

concentrate ('Aatrex 280') and either of two formulations of isazofos, (i) 100 g kg<sup>-1</sup> granules, or (ii) 500 g litre<sup>-1</sup> microencapsulated ('Miral 500') were applied to all the lysimeters. The isazofos 100 g kg<sup>-1</sup> granule was formulated as follows: the granular substrate was 24-48-mesh corn cob granules ('Agsorb 24-48 LVM'). Technical isazofos (93.7 % purity) was sprayed onto the surface of the granules. The granules contained no dust, were resistant to abrasion and did not crumble in water. Microencapsulated isazofos was a water-based suspension of microscopic polymer capsules containing technical grade isazofos at 500 g litre<sup>-1</sup> and polymer at 100 g litre<sup>-1</sup>. Atrazine was applied at the rate of 2.25 kg a.i. ha<sup>-1</sup> (3.98 mg core<sup>-1</sup>)<sup>2</sup>. Isazofos was applied at a rate of 4.25 kg a.i. ha<sup>-1</sup> (7.53 mg core<sup>-1</sup>) for both formulations. The granular formulation was applied 2.5 cm below the soil surface (at least 1.0 cm from cylinder wall) by removing and replacing the top soil layer. Liquid formulations were applied by pipet (10 ml aqueous suspension) to the surface of each soil core in a spiral fashion starting at least 1 cm away from the cylinder wall.

## 2.2 Experimental set-up

### 2.2.1. Field studies

Three sets of 24 stainless steel lysimeters<sup>3,4</sup> (75 cm long x 15 cm diameter, packed with 70 cm of soil) were set up as follows: (i) Set 1-- isazofos granules in Plainfield sand (sand 87.5 %, silt 6.5 %, clay 6.0 %, organic matter 1.5 %, pH 5.55<sup>5</sup>); (ii) Set 2 -- isazofos, microencapsulated in Plainfield sand; (iii) Set 3 -- isazofos, microencapsulated in Vittoria loam (0-15 cm: sand 43.0%, silt 45.5 %, clay 11.5 %, organic matter 4.48 %, pH 5.81; 15-70 cm: sand 82.0 %, silt 11.5 %, clay 6.5 %, organic matter 1.47 %, pH 6.01). In lysimeters using Vittoria loam, the bottom 55 cm were initially packed with Vittoria subsoil which was allowed to settle before adding the top 15 cm of top soil. Atrazine was applied to all lysimeters as an internal reference. Details of experimental procedures have been described previously.<sup>1,3</sup> Approximately 12 litre of water was applied to each lysimeter over the two-week equilibration period preceding pesticide application on 8 May, 1990. Each lysimeter set was divided into two moisture regimes (six pairs each) in which half of the lysimeters received only natural rainfall, while the remaining lysimeters received rainfall plus supplementary watering, simulating a 50-mm rainfall on days 2 and 9. After week 2, 25-mm water applications were made simulating an irrigation protocol: (a) two days following less than 10 mm rainfall, (b) three days following 10 to 25 mm rainfall, (c) four days following > 25 mm rainfall, or the last watering. One pair of 15-cm x 70-cm lysimeters from each moisture regime was retrieved from the field at 1, 2, 4, 8, 12 and 21 weeks, and frozen for subsequent sectioning (seven segments, each 10 cm long) and residue analysis. Effluent was pumped from the 1-litre Pyrex beaker below each lysimeter on the day following each rainfall or watering. Volumes were recorded and sub-samples were centrifuged to remove any particulates, then taken for analysis.

### 2.2.1 Laboratory studies

Laboratory persistence studies for isazofos were conducted using three formulations (emulsifiable concentrate (EC), granule and microencapsulated) and two soils (Plainfield sand (20 % moisture), and Vittoria loam (31 % moisture)). One kilogram of each soil was treated with one of the isazofos formulations, and then divided into two replicates which were sampled at 0, 1, 2, 3, and 8 weeks. Sample containers with plastic sheet covers were weighed at start-up and water was added as required to maintain the respective initial moisture contents. Isazofos application rates were the same as those used in the lysimeter studies, based on the 3.0 kg wet weight (approximate) of the lysimeter segments. Additional samples of the microencapsulated isazofos-Plainfield sand were analyzed at 16 weeks in order to calculate  $DT_{50}$  values (50 % disappearance time). Each sub-sample was

incubated at 22°C in a glass container covered with plastic film to minimize moisture losses. Extraction procedures were the same as those used for soil core extraction.

## 2.3 Soil core extraction and analysis

Soil core sub-samples (100 g) were extracted using HPLC-grade methanol + water (90 + 10 by volume) as reported earlier.<sup>1</sup> Extracts were filtered through vacuum Buchner funnels and filtrates were quantitatively transferred to 250-ml volumetric flasks, made up to volume with water, and centrifuged for 10 min to remove sediment traces before HPLC or GLC analysis.

Isazofos and atrazine analyses were done by GLC, using the following operating parameters: column, J&W DB-17, 0-53 : m x 15 m (megabore, 1.0: m film); carrier gas, ultrapure helium (15 to 20 ml min<sup>-1</sup>); detector, nitrogen-phosphorus, 260°C; hydrogen flow rate, 1 to 3 ml min<sup>-1</sup>; injector temperature, 240°C; injection volume, 2.0: l. Minimum detectable concentrations for isazofos and atrazine were, respectively, 1 : g litre<sup>-1</sup>, and 5 to 10 : g litre<sup>-1</sup>. Atrazine content was confirmed by HPLC analysis. The operating parameters were: column, Waters Resolve column 15.0 x 3.9 mm reversed-phase C18, 3 : m particle size; pump speed, 0-9 ml min<sup>-1</sup>; injection volume, 40 : l; detector wavelength, 220 nm; mobile phase, acetonitrile + water (1 + 1 by volume). The minimum detectable concentration was 5 to 15 : g litre<sup>-1</sup>.

## 2.4 Soil-water partitioning studies

Batch adsorption studies were carried out to obtain Freundlich soil-water partitioning coefficients ( $K_d$ ) for each pesticide-soil combination used in the study.<sup>6</sup> Thirty-millilitre volumes of five different initial pesticide concentrations (ranging from 1 to 20 : g ml<sup>-1</sup>), in triplicate, were added to 8.0-g air-dried samples of the appropriate soil, then tumbled in 120-ml glass bottles for 18 h. Samples were then centrifuged at 40000 g for 0.5 h to separate the phases. Aqueous samples were analyzed by either GLC or HPLC. Adsorption data were plotted in the log-Freundlich format to calculate  $K_d$  values.

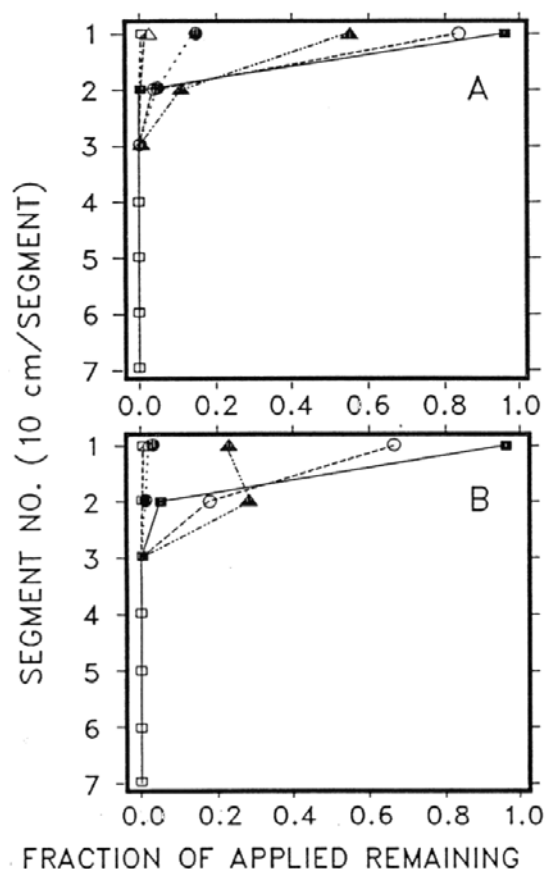
## 3 RESULTS AND DISCUSSION

### 3.1 General

An earlier-reported 1989 isazofos lysimeter study<sup>1</sup> was continued when the microencapsulated formulation of isazofos became available.

**TABLE 1.** Precipitation Received 10 May to 3 October 1989, and 8 May to 2 October 1990, by Lysimeters under both Natural Rainfall and Supplementary Watering Treatments

Week	Cumulative precipitation (mm)			
	1989		1990	
	Rainfall	Watering	Rainfall	Watering
1	6.0	107.7	36.0	86.8
2	23.0	124.7	90.5	19.2
4	95.0	222.1	10.5	261.0
8	133.5	362.3	177.0	431.2
12	149.5	505.4	308.0	587.7
21	226.5	811.2	553.0	883.5



**Fig. 1.** Concentration profiles for granular formulations of isazofos in Plainfield sand. A, rainfall; B, supplementary watering. Week ( ) 1; (O) 2; (•) 4; ( ) 8; (◊) 12; (□) 21.

To facilitate comparison with the earlier study, the isazofos 100 g kg<sup>-1</sup> granule treatment using Plainfield sand was repeated in 1990, using atrazine as an internal reference. While the amounts of supplementary water applied in both years (Table 1) varied by only 4.3 % from the mean, the amount of rainfall received during the 1990 study was more than

double that in 1989. The ability to control moisture inputs in these lysimeter studies by applying supplementary water to selected treatments has proven very useful in making mobility comparisons between data sets from different years, which would not have been possible using conventional field-plot studies relying only on natural rainfall.

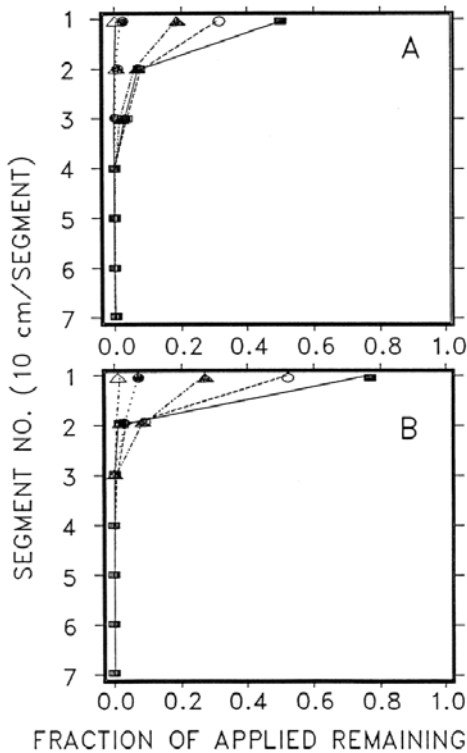
### 3.2 Effluent data

No isazofos was observed in the effluent under either moisture regime from either granular or microencapsulated formulations applied to Plainfield sand cores, or from the microencapsulated formulation applied to Vittoria loam cores throughout the study (1 : g litre<sup>-1</sup> detection limit). Only a single atrazine elution was observed in one Plainfield sand core under supplementary watering, whereas atrazine eluted from about 50 of the Vittoria loam cores.

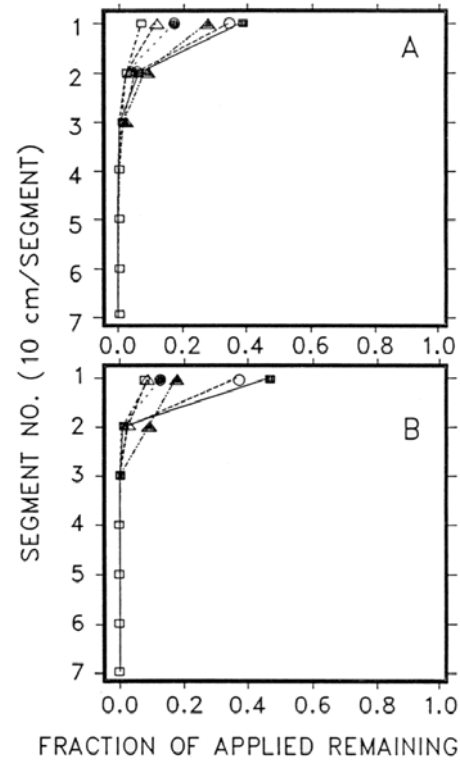
### 3.3 Mobility data

Under rainfall, microencapsulated isazofos was essentially confined to the top 10-cm segment of Plainfield sand cores (concentration profiles for microencapsulated isazofos are not shown because of minimal movement), whereas moderate amounts of isazofos from the granular formulation (11% of applied, Fig. 1A) moved into the second segment (maximum of 20 cm) by week 4. Under supplementary watering, microencapsulated isazofos showed very slight movement into the second segment, with traces also in segment 3, while substantial amounts of isazofos from the granular formulation (29 % of applied) moved into the second segment (Fig. 1B) by week 4. Isazofos from the granular formulation exhibited similar mobility characteristics in Plainfield sand cores in both the current study (Fig. 1) and in the 1989 study (Fig. 2B, adapted from Ref. 1). While more isazofos from the granules moved into segment 2 in the current study (Fig. 1 B), it penetrated further down the soil cores in the 1989 study (Fig. 2B). Isazofos from the EC formulation in the 1989 study (Fig. 2A, adapted from Ref. 1) exhibited much greater mobility than it did from the granular formulation. Traces of isazofos reached the base of the lysimeter after one week, with 22 elutions of isazofos being recorded from lysimeters in that treatment throughout the study.<sup>1</sup>

Microencapsulated isazofos in the current study exhibited greater mobility in the heavier-textured Vittoria loam (Fig. 3) than in Plainfield sand, where there was minimal movement. This mobility behaviour was not consistent with the respective Freundlich adsorption coefficients,  $K_d$  (Table 2), which were obtained using analytical-grade chemicals. Based on the  $K_d$  values for isazofos of 0.843 and 1.929 : g<sup>(1-N)</sup> g<sup>-1</sup> ml<sup>N</sup> for Plainfield sand and Vittoria loam (surface), respectively, one might expect isazofos to be less mobile in the Vittoria loam soil. However, similar increased pesticide mobility in heavier-textured soils was previously observed in Honeywood silt loam



**Fig. 2.** Concentration profiles for isazofos formulations in Plainfield sand, under supplementary watering in 1989. **A**, EC formulation; **B**, granular formulation. Week ( ) 1; (O) 2; (•) 4; ( ) 8; ( ) 12; (□) 21.



**Fig. 3.** Concentration profiles for a microencapsulated formulation of isazofos in Vittoria loam. **A**, supplementary watering; **B**, rainfall. Week ( ) 1; (O) 2; (•) 4; ( ) 8; ( ) 12; (□) 21.

**TABLE 2**

Freundlich Adsorption Constants ( $K_d$ ,  $N$ )<sup>a</sup> for Atrazine and Isazofos Adsorption by Plainfield Sand and Vittoria Loam

Soil	Atrazine		Isazofos	
	$K_d^a$	$N^b$	$K_d$	$N$
Plainfield sand	0.843	0.774	1.426	0.889
Vittoria loam-Top	1.929	0.810	2.663	0.861
Vittoria loam-Sub	0.486	0.925	1.047	0.894

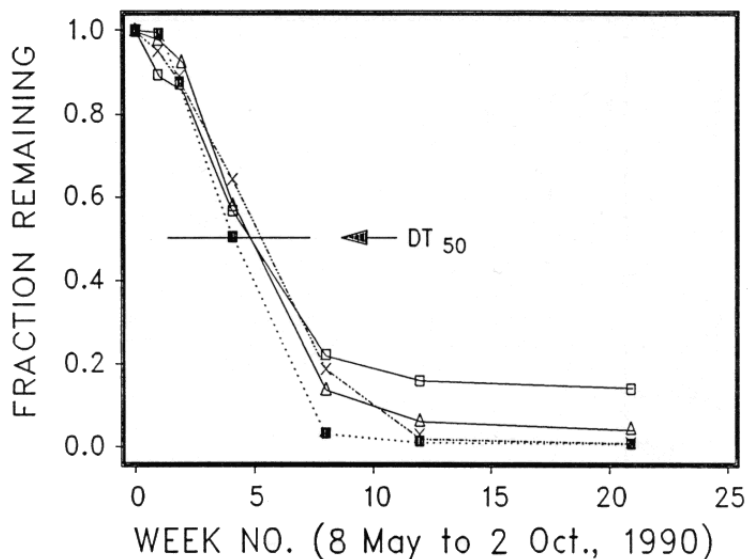
<sup>a</sup>  $K_d$  = adsorption coefficient; :  $g^{(1-N)} g^{-1} ml^N$ .  
<sup>b</sup>  $N$  = Freundlich isotherm slope.

cores treated with isazofos EC<sup>3</sup> or terbutylazine<sup>4</sup> (N<sup>2</sup>-tert-butyl-6-chloro-N<sup>4</sup>-ethyl-1,3,5-triazine-2,4-diamine), a triazine herbicide. In both instances, the heavier-textured soils were packed into the lysimeters in two stages, the lower 55 cm of coarser subsoil underlying the top 15 cm of finer-textured top soil.

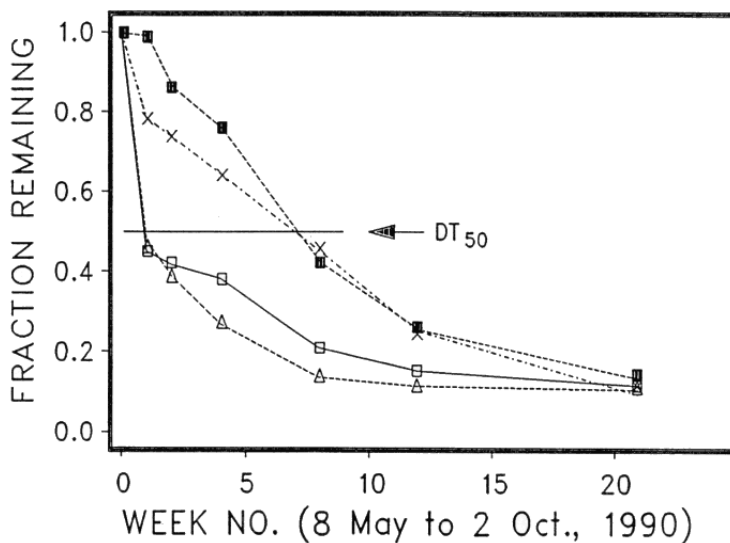
According to Hillel and Baker,<sup>7</sup> unstable water flow (streaming or fingering) can occur in soil profiles where a more highly conducting coarser horizon underlies a finer horizon, and where surface ponding exists, as was the case here. This fingering phenomenon is enhanced whenever the sublayer's water entry is very low (controlled by the surface horizon hydraulic conductivity) while having a very high hydraulic conductivity. Much of the pesticide transport occurred following the 50-mm water application on the day after pesticide application, in which the water remained ponded much longer on the surface of the heavier-textured soils. Similar scenarios could occur under actual field situations, when a heavy rain producing ponding occurs following chemical application on a depressional area of a silty loam surface soil, underlain by a coarser subsoil.

**3.4 Persistence data**

We report persistence data in terms of observed 50 disappearance times (DT<sub>50</sub>) rather than calculating simple first-order decay constants, since several disappearance curves had distinctly different kinetics operating at different times throughout the study.



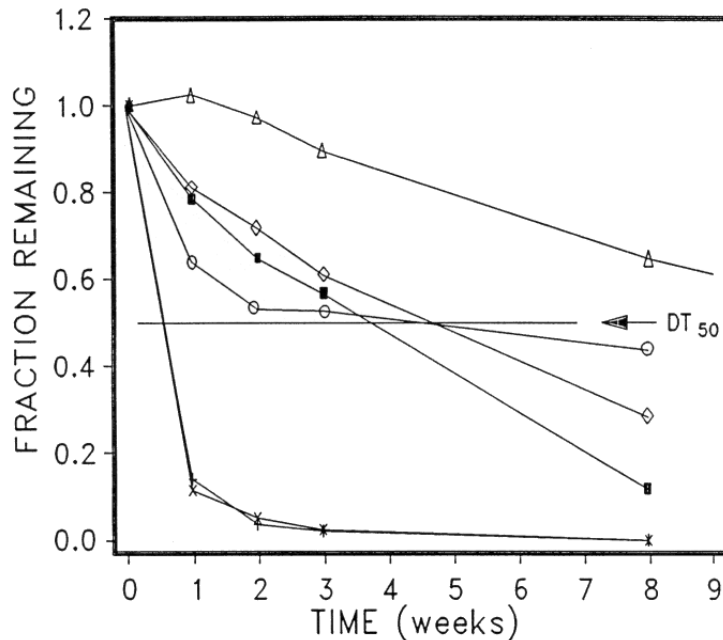
**Fig. 4.** Isazofos persistence in Plainfield sand, as influenced by formulation. (□) Microencapsulated, water ( $DT_{50}$ : 4.8 week); (○) microencapsulated, rain ( $DT_{50}$ : 4.8 week); (△) granular, water ( $DT_{50}$ : 4.1 week); (x) granular, rain ( $DT_{50}$ : 5.3 week).



**Fig. 5.** Persistence of atrazine and microencapsulated isazofos in Vittoria loam, under rainfall and supplementary watering. (□) Isazofos, water ( $DT_{50}$ : 0.94 week); (○) isazofos, rain ( $DT_{50}$ : 0.94 week); (△) atrazine, water ( $DT_{50}$ : 7.1 week); (x) atrazine, rain ( $DT_{50}$ : 6.9 week).

In fact, kinetics may well have followed first-order behaviour at the microscale, but may have been controlled by spatially and temporally variable factors such as moisture, temperature, or nutrient status, which varied throughout the study<sup>8</sup>.  $DT_{50}$  values for

microencapsulated isazofos in Plainfield sand were 4.8 week for both moisture regimes (Fig. 4). The  $DT_{50}$  of granular isazofos was 5.3 week under rain, but only 4.1 week under supplementary watering.



**Fig. 6.** Laboratory persistence of isazofos formulations in Plainfield sand, at 20% moisture: ( ) granular,  $DT_{50}$ : 4.6 week; ( ) microencapsulated,  $DT_{50}$ : 12.0 week; ( ) EC,  $DT_{50}$ : 3.8 week; and Vittoria loam, at 31 % moisture: (+) granular,  $DT_{50}$ : 0.53 week; (O) microencapsulated,  $DT_{50}$ : 4.2 week; (x) EC,  $DT_{50}$ : 0.53 week.

By week 12, granular isazofos had virtually disappeared under both moisture regimes, whereas 5 and 16% of the microencapsulated isazofos remained at week 21 under rain and supplementary watering treatments, respectively.

The microencapsulated and granular formulations of isazofos had similar disappearance rates in Plainfield sand cores (Fig. 4) through the first eight weeks of the study, after which the disappearance rate from the microencapsulated formulation slowed abruptly. In the 1989 study,<sup>1</sup> the granular formulation was more persistent than the EC formulation in the same Plainfield sand as used in this study. The changing disappearance rates for the formulated products may reflect the protection from microbial action afforded by the formulation.

More than 50 % of applied microencapsulated isazofos disappeared from Vittoria loam cores within one week (Fig. 5) under both moisture regimes ( $DT_{50}$  was 0.94 week for both), while  $DT_{50}$  values for atrazine were nominal at 7.1 and 6.9 week for supplementary watering and rainfall treatments, respectively. For this reason, isazofos concentrations shown at week 1 in Fig. 3A and 3B were much lower than were observed in Plainfield sand. A series of extraction recovery tests was conducted for microencapsulated isazofos on this soil at different moisture contents and using variations of the extraction procedure, without improving recoveries.

In the laboratory persistence study, we used all three isazofos formulations and both soils, maintaining approximately the same soil moisture contents as were measured in the field lysimeters. All zero day recoveries exceeded 95 % of applied amounts.

Isazofos from both EC and granular formulations disappeared very quickly from Vittoria loam ( $DT_{50}$  = 0.53 week, Fig. 6) while the microencapsulated isazofos disappearance rate was initially rapid, but then slowed considerably after two weeks ( $DT_{50}$  = 4.2 week). Approximately 35 % of the microencapsulated isazofos was immediately available for degradation during the first week, while the remainder may have been protected by the formulation, resulting in a slower degradation rate thereafter (Fig. 6).

### 3.5 Atrazine-isazofos interaction

Atrazine has been applied to all soil cores in our lysimeter studies for several years as an internal reference, facilitating mobility and persistence comparisons among treatments of different years. In 1989, we observed that the presence of granular isazofos appeared to extend atrazine persistence in Plainfield sand under rainfall (relative to atrazine alone or in combination with other herbicides). Atrazine persistence data from that study appear in Table 3 along with additional data from this study.

**TABLE 3**

DT<sub>50</sub> (Time for 50 % Disappearance) Values for Atrazine in Plainfield Sand Lysimeters as Affected by Co-application of Metribuzin, or Various Isazofos Formulations, in 1989 and 1990.

Treatment	DT <sub>50</sub> atrazine (week)	
	Watering	Rainfall
	1989	
Metribuzin	5.1	4.9
EC isazofos	5.5	5.8
Granular isazofos	6.0	7.3
	1990	
Microencapsulated isazofos	5.4	6.8
Granular isazofos	6.7	9.4

If one hypothesizes that isazofos must be in close proximity to atrazine for it to influence atrazine degradation, then this condition was best satisfied for the longest period by the more persistent isazofos granular formulation under rainfall, in which minimal movement of both atrazine and isazofos occurred. EC isazofos might be expected to have the least effect upon atrazine degradation since it was less persistent and leached further from the application point (Fig. 2), reducing the average soil concentration. In the 1989 study, DT<sub>50</sub> values for atrazine appeared to be influenced by different combinations of moisture regime (responsible for pesticide movement) and by isazofos formulation. Atrazine DT<sub>50</sub> values were 4.9 and 5.1 week and 5.5 and 5.8 week, for the metribuzin-atrazine and EC isazofos-atrazine combinations under watering and rainfall, respectively (Table 3). If, according to our hypothesis, we consider the metribuzin- and EC isazofos-atrazine combinations (four values) as 'no-effect' levels, then the mean DT<sub>50</sub> was 5.3 week, with a standard deviation of ± 0.4 week. In contrast, the atrazine DT<sub>50</sub> value (atrazine-isazofos granules, rainfall) was 2.0 week greater at 7.3 week, and 13 week greater than the highest DT<sub>50</sub> value in the 'no-effect' group (5.8 week, EC isazofos (rainfall)).

In the current study, atrazine persistence again appeared to be influenced by both isazofos formulation and moisture regime (Table 3). The combination of the granular formulation and the rainfall treatment again produced the largest atrazine DT<sub>50</sub> value at 9.4 week, 2.7 week and 4.0 week longer than in the granular and microencapsulated treatments (watering), respectively. Atrazine DT<sub>50</sub> values from the isazofos granular treatment (both moisture regimes) were somewhat greater than observed in 1989, perhaps due to differing weather conditions between the two years.

Two possible scenarios for this interaction are: (1) isazofos inhibited microorganisms responsible for degrading atrazine, thereby extending its persistence; or (2) an isazofos metabolite, such as the hydrolysis product CGA17193 (5-chloro-1-isopropyl-1H-1,2,4-triazol-3-ol), might have inhibited atrazine degradation, although it is very mobile<sup>1</sup> and would have quickly been

transported away from the target atrazine by any significant water flow. Admittedly, its mobility would have been minimized in the rainfall-only treatment. In the current study, atrazine in the granular isazofos application-rainfall treatment showed negligible degradation after week 2, and only 15 % disappearance by week 4. In comparison, atrazine under the other isazofos-moisture combinations decreased, on average, by 4, 8 and 28 % at weeks 1, 2 and 4, respectively. During this initial two-week period, less than 10% of applied isazofos had disappeared (Fig. 4), reducing the plausibility of scenario 2, unless the small amount of metabolite produced had a very potent effect upon microbial degradation of atrazine. After the initial two-week inhibition, atrazine disappearance rates in the granular isazofos treatment paralleled disappearance rates under the other treatments.

It is important to emphasize that although we have considerable confidence in these atrazine persistence data (each of six data points making up a degradation curve came from a different pair of lysimeters-12 lysimeters per degradation curve (i.e. Figs 4, 5)), this facet was not an original objective of the study. However, these observations appear to be of importance in a retrospective analysis and may be useful in designing future replicated field microplot-on-pesticide interactions.

#### 4 CONCLUSIONS

In summary we report the following significant findings in this study:

1. Isazofos mobility from the three formulations in this and the previous study<sup>1</sup> increased in the following order in Plainfield sand: microencapsulated < granular < EC.
2. Isazofos from the microencapsulated application was more mobile in the heavier-textured Vittoria loam than in Plainfield sand, following the 50-mm water application on day 2. Conditions appeared appropriate (extended ponding, textural change between horizons) for fingering or unstable water flow to occur, resulting in rapid solute movement. This mobility behaviour was not consistent with the respective adsorption tendencies indicated by the Freundlich adsorption coefficients.
3. Observed atrazine persistence in Plainfield sand from the previous<sup>1</sup> and current studies suggested that isazofos (granular) decreased the atrazine disappearance rate during initial stages of the study. By week 4 in this study, the atrazine disappearance rate in this treatment paralleled its disappearance rate in other treatments.
4. The rapid isazofos disappearance rate from the microencapsulated formulation in Vittoria loam during the first week following application suggested that the microbial population was able

to degrade isazofos with little or no lag period. In laboratory persistence studies using this soil, even higher disappearance rates were observed for EC and granular formulations of isazofos. Much slower disappearance rates were observed for all three formulations in Plainfield sand.

#### ACKNOWLEDGEMENTS

The author is grateful for both financial and product support furnished by Ciba-Geigy, USA for this study, and for helpful suggestions of Dr John Purdy, Ciba Geigy, Canada. The author wishes to record his appreciation of the able technical assistance of Pat Moy, Susan Sibbald, and LouAnn Verellen in collecting and analyzing the data used in preparations of this manuscript.

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