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# Effect of Spatial Heterogeneities of Water Fluxes and Application Pattern on Cadusafos Fate on Banana-Cultivated Andosols

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In tropical humid environments under intensive banana production, pesticide transfer in waters can be of particular concern due to heavy rainfall, steep slopes, and soils with high infiltration capacities. The transfer in percolation and runoff waters of the nematicide cadusafos was investigated during a three month field experiment. The spatial heterogeneity of the banana plantation was taken into account by measuring percolation fluxes both under the banana plants and in the interrows with a specially designed lysimeter device installed at 60 cm depth. At the field scale, 0.34% of the pesticide applied was transferred in percolation, 0.13% in runoff. Forty-nine percent of cadusafos losses occurred by percolation under the banana plants, 23% by interrow percolation, and 28% by runoff. Losses were highest during the three weeks following cadusafos application, and this is also when dissipation in the soil was highest (calculated half-life in the soil: 7d). After this period, losses of cadusafos were low, both in soil and waters. Under the banana plant, saturated fluxes carried most of the pesticide, despite total percolation fluxes being at least five-times higher than saturated ones. Although overall pesticide transfer in water was low (0.5% of applied), it was not negligible due to the frequency of pesticide application in these areas.

KEYWORDS: Runoff; infiltration; lysimeters; andosols; pesticide; banana plantations; French West Indies

# INTRODUCTION

Environmental surveys have demonstrated that the aquatic environments of the Caribbean regions and Central America, as well as agricultural soils from these areas, are widely contaminated by a suite of pesticides (1-5). These regions show particular factors: steep slopes; heavy rainfalls, e.g., 4000 mm annually in Guadeloupe (French West Indies) (6); soils with high infiltration capacities; heterogeneous land cover with various crops which include extensive uses of pesticides. The combination of these factors represents a worst-case scenario for contamination of surface, coastal, and subterraneous waters by pesticides (7). This highlights the need for a better understanding of the pesticide fate in these conditions in order to seek new management strategies that could limit the contamination of water resources.

The main paths for water contamination by pesticides are dependent on soil, climate, and crop (8). In addition, pesticide

losses are variable with time. In many instances, it has been shown that the first storm events after application are responsible for most of the seasonal water contamination and that transport by surface runoff is the main process by which water contamination occurs (9, 10). For example, in vineyards in the Mediterranean area where periods of drought are followed by intense rainfall, losses by surface runoff during the first storm event can represent more than 80% of the seasonal losses of pesticide molecules to surface waters (11). Most of these observations have, however, been made in temperate or semiarid environments. Climatic conditions are different in humid tropical environments: rainfall is much higher and occurs regularly throughout most of the year, which often leads to a situation of soil saturation and permanent percolation fluxes (12) and overland flow (13). Consequently, percolation as well as surface runoff may be of importance for pesticide transfers, and losses may be more regular than in other climates. Furthermore, given high soil saturation conditions, percolation fluxes may be of a different type, including preferential flow as demonstrated in tropical volcanic soils (14, 15). These preferential flows may enhance pesticide losses since they are generally associated with preferential fluxes of pollutants (16-19). Finally, for tropical conditions, there is a lack of knowledge about the relative

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contributions of overland flow and percolation fluxes to pesticides losses.

Agricultural practices are one of the main determinants of pesticide loss rates. This is particularly the case for crops with high inputs of pesticides like banana plantations. Henriques et al. (20) listed not less than 26 active ingredients used on this crop in Latin America while the United Nations Environment Programme estimates that 75-250 kg active ingredient (a.i.) ha<sup>-1</sup> year<sup>-1</sup> of pesticides are applied in banana plantations in Panama, and 36 kg a.i.  $ha^{-1} year^{-1}$  in Costa Rica (21). In the French West Indies, pesticides are applied at different times, typically from February to March, from May to June, and from September to October (22). As a result, water analyses from the "Direction Régionale de l'Environnement" in the French West Indies showed the presence of pesticide residues, such as the nematicide cadusafos, in catchment outlets and groundwater with concentrations exceeding in some cases the threshold for potable water (0.1  $\mu$ g L<sup>-1</sup>). These pesticide residues interact with the hydrological functioning of the cultivated crop. Given (i) the spatial organization in rows and interrows, (ii) the cropping practices leading to anisotropic soil physical properties (23, 24), and (iii) the particular shape of banana leaves, banana plantations exhibit a complex water flux system. Indeed, the rainfall redistribution by banana plant leads to significant stemflow, which cause a local increase in rainfall amount and intensity at the base of the plant (25). This favors percolation fluxes in this location and more specifically gravity fluxes (26) as well as leaching of fertilizers applied at this location (27). In addition, it also favors surface runoff which can represent between 10 to 35% of rainfall during a runoff event (28) and could thereby greatly increase the risk of pesticide losses. Finally, the risk of water contamination by pesticides due to the hydrological behavior of banana canopies seems maximal around the banana plant, and this may be enhanced by the fact that many pesticides used for banana crop protection are applied at the base of banana plants to ensure a maximum effect against root parasites. However, as far as we know, no observation has confirmed the assumption that the spatial heterogeneities due to cropping patterns in banana plantations influence pesticide transfer.

The fate of pesticides is also highly dependent on soil properties. In tropical volcanic conditions, andosols cover large agricultural areas. They show a high content in minerals such as allophanes which, due to their important specific surface area, have an enhanced capacity to bind organic matter. In these conditions, Sansoulet et al. (27) showed that potassium and nitrate leaching was retarded because of the particular cationic and anionic soil capacity, but only a few studies have investigated the sorption behavior of pesticides in andosols (29-31).

Our study therefore aimed to assess the effect of spatially heterogeneous cadusafos application and hydrological functioning on the losses of this pesticide in a banana plot in a tropical volcanic environment, in Guadeloupe. More precisely, we aimed to quantify the contribution of percolation fluxes and runoff to the losses of cadusafos to enable better assessment of surface and groundwater contamination processes. The elucidation of the influence of spatial cropping patterns on water contamination should provide additional information for improving agricultural practices in order to better preserve the quality of water resources in tropical areas where banana plantations are major agricultural enterprises.

#### MATERIALS AND METHODS

**Study Site.** The study was conducted at the Neufchâteau research station (16°04'38" N, 61°36'04" W, 250 m), on the windward side of Basse Terre, Guadeloupe. Mean annual rainfall at the research station is 3850 mm (6). The field experiment was located at the field site "Espérance-Haut" described in Field Experimental Setup.

The soil type is an umbric andosol (32) that has already been described in Cattan et al. (28). Briefly, the A horizon (0–30 cm) has a loamy texture, contains 60 g kg<sup>-1</sup> of organic carbon. The undisturbed B horizon (30–60 cm) has a fluffy texture typical for allophane soils, and a water content that never falls below 1000 g kg<sup>-1</sup> under field conditions. Its organic carbon content is 30 g kg<sup>-1</sup>. It has a continuous macroscopic structure, with medium and fine tubular pores.

**Pesticide.** Cadusafos [*S*,*S*-di-*sec*-butyl *O*-ethyl phosphorodithioate] is an organophosophorus nematicide. It was applied as a granular formulation with 10% a.i. (Rugby 10 G FMC Corp., Philadelphia, PA). Cadusafos controls various nematodes and larvae of *Noctuidae*, *Agriotes* spp. and other soil insects. Its recommended application rate is 3 to 10 kg a.i. ha<sup>-1</sup> (*33*). The water solubility of cadusafos is 245 mg L<sup>-1</sup>, its dimensionless Henry's constant (*K*<sub>h</sub>) is  $5.42 \times 10^{-5}$ , its vapor pressure is 119.6 mPa, and it has a  $K_{oc}$  of 227 mL g<sup>-1</sup> OC.

The application of Rugby 10 G was performed on September 20, 2001, on the experimental field at a rate of 6 kg of a.i.  $ha^{-1}$ . Given the banana plant density of 1810 plants  $ha^{-1}$ , 33 g of Rugby 10 G was applied evenly around the pseudostem at the base of each banana plant on the whole field. No Rugby 10 G had been applied since June 2000.

**Field Experimental Setup.** The field "Espérance Haut" has a 6000 m<sup>2</sup> surface area and a mean slope of 12%. The previous crop was banana, followed by 8 months of fallow. Banana was planted on February 21, 2001, in a square block design ( $2.35 \times 2.35$  m; 1810 plants ha<sup>-1</sup>) (**Figure 1a**). Rainfall volume and intensity were measured on site using a rain gauge (ARG100, Campbell Scientific, Shepshed, Leicestershire, U.K.) with a tipping bucket having a measurement resolution of 0.2 mm, with a 1-min time lapse.

For the purpose of another experiment conducted on the same site to study runoff, the field "Espérance-Haut" had been divided into two subplots with different managements of the banana plantation (in terms of banana planting and soil tillage) (28). However, this layout was not appropriate to study the effect of the type of management practices on pesticide losses: on the one hand, the accuracy was not sufficient to show an effect on percolation fluxes; on the other hand, there were no replicates for runoff. In the present study, we therefore considered the two plots as replicates. These two subplots were separated by 50 cm wide galvanized sheets pushed vertically 20 cm into the ground. The runoff from each plot was channeled to the outlet via a concrete-lined channel at the lower end of the subplot and then to a venturi channel (type E 1253 AZ, Hydrologic, Grenoble, France). The head of water in each venturi channel was measured using a bubble flowmeter (ALPHEE 3010, Hydrologic, Grenoble, France) adapted to the narrow width of the venturi channel, with an 8-s time lapse. Each venturi channel was equipped with a sampling machine controlled by the flowmeter. The samplers (sampler 900 MAX, American SIGMA, Loveland, CO) had 12 glass bottles of 1 L each.

For percolation fluxes, two different sites were selected for lysimeter installation: the base of banana plants and the space between banana plants (Figure 1b). These two positions were chosen to account for the heterogeneity in the rainfall distribution in the banana plantation, spatial variations in soil infiltration due to agricultural practices (tillage, planting techniques, etc.), and the heterogeneity in the distribution of pesticide applications. Indeed, the base of banana plants is an area where rainfall accumulates after stemflow, and it is also where fertilizers and pesticides are mostly applied. The area between banana plants is generally protected by the canopy from incident rainfall. On the basis of these factors, for percolation measurement, the experimental setup included eight lysimeters installed at four locations on the plot. At each location, one lysimeter was installed directly under a banana plant and another one between plants (Figure 1b). To install the lysimeters in the field, we dug trenches along the field slope beside a banana row and then dug two excavations horizontally at 60 cm in depth in the B horizon, one from the edge of the trench below a banana plant and the other from the uphill edge of the trench. The lysimeters were inserted



Figure 1. (a) Schematic representation of the field "Espérance-Haut" and the experimental layout. (b) Diagram indicating the position of lysimeters in the banana field, the zone of application of pesticide, and the soil sampling sites.



Figure 2. Schematic diagram of a lysimeter for wick and gravity sampling (after Cattan et al. (34)).

in the excavations. Finally, the lysimeters were wedged with wooden blocks and the trench was backfilled with the excavated material.

The lysimeters used consisted of a  $75 \times 75$  cm galvanized iron plate of 2 mm thickness (Figure 2), containing three  $20 \times 20$  cm compartments located in the middle of the main plate and designed to collect water via fiberglass wicks. The wicks enabled imposition at the soil/lysimeter interface of a negative tension, ranging from 0 to -5 kPa according to flow conditions. The fiberglass wicks were 1.27 cm diameter (1/2" diameter round fiberglass, Pepperell Braiding Company, MA) with a saturated hydraulic conductivity of 11680 mm  $h^{-1}$ . The wicks, each 50 cm long, were unbraided, frayed, and spread across soil aggregates scattered on the surface of each compartment to ensure contact with the soil overlying the sampler. In each compartment, the wicks were inserted in a hole that was drilled to channel the sampled water to underlying collection chambers. The rest of the plate served as a gravity lysimeter designed to collect percolated water samples with the soil/lysimeter interface set at atmospheric pressure. As described in Cattan et al. (34), using simultaneously gravity and wick lysimeters allows estimation of boundary values of the actual percolation fluxes and differentiation between the contributions of saturated and unsaturated fluxes to percolation, since gravity lysimeters sample only saturated fluxes whereas the wick lysimeters sample both types of fluxes.

**Measurements.** The experiment lasted from September 17, 2001, to December 15, 2001. For runoff water, the sampling program began when the water height in the venturi channel was more than 10 mm. Then one sample of 1 L was taken when runoff volume since last sampling became larger than 45 L and the delay since the last sampling exceeded 1 min. Since most rainfall events had a short duration (median value of 10 min according to Cattan et al. (25)), these conditions allowed sampling of most runoff events, even the small ones (less than 0.15 mm). Each day, the sampling bottles for each runoff event were mixed and a mean sample was built up per runoff event. However, owing to the limited number of bottles and to the high number of rainfall events per day (median value of 3 according to Cattan et al. (25)), we could not analyze all runoff events in terms of pesticide concentration. Thirty-

eight runoff events were analyzed for cadusafos concentration including one sample before pesticide application.

For percolation fluxes, we obtained a total of 58 measurements per lysimeter. The frequency of sample was on average three times a week. The frequency increased during certain periods in order to monitor and measure more closely cadusafos concentrations in the percolation waters. Similarly, the frequency decreased during periods without rainfall. Both runoff and percolation waters were filtered using glass Fiber Filters (4.7 cm diameters, 0.45  $\mu$ m porosity, Whatman GF/C), but no suspended soil material was measured. Water samples were frozen at -18 °C for a maximum of two weeks before analysis at the Laboratory of INRA at Arras, France.

For each subplot of 3000 m<sup>2</sup>, we collected one bulk sample at three different depths (0-5, 5-30, 30-60 cm) and three different positions to account for the spatial heterogeneity of cadusafos application: in the interrow, at 30 cm upstream of the banana plant and at 30 cm downstream of the banana plant, as shown in Figure 1b. Each bulk sample representative of a given depth and position was a mix of four samples spatially distributed in the subplot. Soil samples were collected with a hand auger of 8 cm diameter which allowed collection of approximately 250 g of soil. Therefore, each bulk sample weighed approximately 1 kg. During the experiment, one sample was collected just before the pesticide application, four samples during the four weeks after pesticide application at the rate of one sample a week and three samples during the last two months of the experiment at the rate of one sample every three weeks. Soil samples were weighed and frozen at -18 °C for a maximum of two weeks before analysis at the Laboratory of INRA at Arras, France. In the expression of results, given the fact that cadusafos was applied homogeneously around the banana plant, we considered that both "upstream" and "downstream" of the banana plant were representative of the same position, namely "under the banana plant". Hence, in the expression of results, n = 2 in the interrow, and n = 4 under the banana plant.

**Cadusafos Analysis in Soil and Water Samples.** The equivalent of 20 g of dry soil was sampled for cadusafos extraction. The extraction was performed using Accelerated Solvent Extraction (ASE 200 Dionex, Sunnyvale, CA) with acetone, at 80 °C, 100 bar for 5 min. Following ASE extraction, partial rotative evaporation was performed. Then, evaporation to dryness was performed under light nitrogen flux. The dry residue was solubilized in 2 mL of hexane and analyzed with gas chromatography (GC). The recovery of cadusafos from spiked samples was 71% for the horizon A, and 40% for the horizon B.

Water samples were stored at 4 °C for a maximum of two weeks before extraction and analysis. The extraction of cadusafos was performed via solid-phase extraction (SPE) with the Autotrace Zymarck automated device (Hopkinton, MA) set up with C18 Alltech cartridges (6 mL, 500 mg). Two-hundred milliliters of aqueous sample were extracted. The elution of the sample was performed with ethyl acetate and dichloromethane. The sample was evaporated under a light nitrogen flux. The residue was diluted in 2 mL of hexane. The quantification of cadusafos from both soil and water samples was done with a gas chromatograph (GC). The extraction efficiency was 86.3%.

GC analysis was performed with a Varian 3400 GC (Palo Alto, CA). The instrument was set up with a split/splitless injector, a thermoionic detector (TSD), and a Restek RTX 200 column (15 m, 0.53 mm, 1  $\mu$ m). The vector gas was helium flowing at 2 mL min<sup>-1</sup>. The temperature of the TSD and the injector were 290 and 260 °C, respectively. The temperature gradient of the oven was from 150 for 1 min and then increasing to 250 °C at a rate of 7.5 °C min<sup>-1</sup> and maintained at this temperature for 2 min. The retention time for cadusafos was 6.05 min. The limit of detection for soil and water samples was 0.5  $\mu$ g kg<sup>-1</sup> soil and 0.01  $\mu$ g L<sup>-1</sup>, respectively.

**Calculations.** For runoff events that were not sampled, we assigned the cadusafos concentrations of the nearest runoff event. We calculated the mean concentration in runoff water for the whole experimental site by averaging the observed data at the outlets of the two plots. For each runoff event, the concentration was multiplied by the volume of water in order to calculate the losses of cadusafos in runoff.

For analyzing the spatial variation of the losses of cadusafos in percolation water, we ran a two-factor analysis of variance (AOV) with four replications on the total losses throughout the period of experiment.  
 Table 1. Comparison of Losses of Cadusafos and of Percolation Volumes in the Wick (WL) and Gravity (GL) Lysimeters under Banana Plant and in the Interrow

	losses (µg m <sup>-2</sup> )			percolation volumes (L m <sup>-2</sup> )		
1st term vs 2nd term	1st term	2nd term	pª	1st term	2nd term	р
Banana WL vs Interrow WL Banana GL vs Interrow GL WI Banana vs GL Banana	13548 12767 13548	739 62 12767	0.09 0.00 0.89	3830 561 3830	1508 61 561	0.18 0.00 0.01
WL Interrow vs GL Interrow	739	62	0.06	1508	61	0.00

<sup>a</sup> Probability of the Tukey's 'Honest Significant Difference' method between the first and second terms, at a confidence level of 0.95. The test was performed on the log values of losses and percolation volumes. The residual term of the analysis of variance is 0.56 for the losses and 0.27 for the volumes.

The two factors considered were (1) the position within the banana field with two levels, under the banana plant or in the interrow, and (2) the kind of percolation measurement with two levels, wick or gravity lysimeters, WL and GL, respectively. Given the skewed distribution of the observed cadusafos losses, the AOV was conducted on the log-transformed values. The test of Tukey was used to compare the means values.

For computing the total percolation losses at the field scale, we distinguished the losses occurring under the banana plants from those occurring between the banana plants. To this aim, we divided the elementary banana planting grid cell of size  $5.522 \text{ m}^2$  (=  $2.35 \text{ m} \times 2.35 \text{ m}$ ) into an "under banana" area equal to  $0.562 \text{ m}^2$  ( $0.75 \times 0.75 \text{ m}$ ) which is the collecting area of one lysimeter and a "between banana" area corresponding to the remaining area of  $4.96 \text{ m}^2$ . It was then assumed that the average fluxes observed on the four lysimeters located under the banana plants and on the four lysimeters installed at the interrows were representative of the "under banana" percolation fluxes and of the "between banana" fluxes, respectively. Eventually, the total percolation losses under and between banana per surface of the banana field were computed as follows:

$$L_{ub} = F_{ub} \times 0.562 \times 1086$$
  
 $L_{bb} = F_{bb} \times 4.96 \times 1086$ 

where  $L_{ub}$  and  $L_{bb}$  were the cadusafos losses by percolation, expressed in g ha<sup>-1</sup> under and between banana,  $F_{ub}$  and  $F_{bb}$  were the estimated average cadusafos fluxes under and between banana, expressed in g m<sup>-2</sup>, and 1086 corresponds to the density of banana plants on 6000 m<sup>2</sup>.

#### RESULTS

**Cadusafos Losses in Percolation Water: Effect of Spatial** Heterogeneity. At the scale of the lysimeter, Table 1 shows that the losses of cadusafos under the banana plant were much larger than in the interrow for the whole period of monitoring. For WL the cumulated amounts in four months reached 13548  $\mu g m^{-2}$  under the banana plant, while in the interrow, losses were 1 order of magnitude less and reached 739  $\mu$ g m<sup>-2</sup>. The losses in GL were of the same order of magnitude as WL under the banana plant (12 767  $\mu$ g m<sup>-2</sup>), but in the interrow they were 10-fold lower in GL compared to WL (62  $\mu$ g m<sup>-2</sup>). These differences between the banana plant and the interrow, irrespective of the type of lysimeter, were consistent with the characteristics of banana plantation. First, because the stemflow process locally concentrates fluxes (25), the percolation fluxes were higher under the banana plant than in the interrow (**Table 1**). Second, the pesticide was applied at the base of the banana plant and this, combined with higher water fluxes, consequently increased the losses at this location. However, there were also some losses in the interrow, meaning that cadusafos was transported from the banana plant area to the interrow. This



**Figure 3.** Concentration of cadusafos in the total percolation (WL) and gravity percolation (GL) under the banana plant during the experiment. Regressions (full line for GL and dashed line for WL) were performed between pesticide application and October 16.

may have occurred either by surface runoff or by lateral fluxes in the soil matrix due to local hydraulic discontinuities caused by a drop in hydraulic conductivity between two soil horizons (135 and 32 mm  $h^{-1}$  for horizons A and B, respectively).

Table 1 also shows that the contribution of saturated fluxes to the pesticide losses was far higher under the banana plant than in the interrow: losses in GL accounted for 94% (12767/ 13548) of the total losses (those from WL) under the banana plant but for only 8% (62/739) in the interrow. This can be related to a higher contribution of saturated fluxes to the total fluxes under the banana plant than in the interrow: they accounted for 15% (561/3830) of the total fluxes under the banana plant and 4% (61/1508) in the interrow. However the difference of contribution of saturated flow to cadusafos percolation losses between the banana plant and the interrow (94% vs 8%) was not proportional to the difference of its contribution to water percolation (15% vs 4%). This suggests either a more than proportional effect of saturated flow on the mobilization of the pesticide molecule or a larger availability of the molecule under the banana plant. This is now analyzed under the banana plant where the losses were maximum.

**Cadusafos Concentration and Losses in Percolation Fluxes** under the Banana: Effect of Time after Application and Type of Percolation. First, we analyzed the concentrations in percolation water and the losses of cadusafos with time. Concentration and losses were maximum immediately after application and decreased rapidly. Concentrations decreased with time in a two-phase trend in both types of lysimeters (Figure 3). The decay was rapid during the first three weeks from the application date till October 16: from 15 to 0.2  $\mu$ g L<sup>-1</sup> in the WL and from 1000 to 0.6  $\mu$ g L<sup>-1</sup> in GL. After this date, and for the next 8 weeks, the concentrations in the two types of lysimeters were constant and of the same order of magnitude, ranging between 0.1 and 0.8  $\mu$ g L<sup>-1</sup>. Despite the fact that rainfall was variable during these weeks, with very humid periods alternating with drier ones (between November 11 and December 1), this did not seem to affect the concentration of cadusafos in drainage fluxes as they remained low. Losses of cadusafos were consistent with the pattern of concentration. For instance,



Figure 4. Ratios of losses and percolation volume between GL and WL under banana plants.

 Table 2. Losses of Cadusafos and Water Drainage by Decades in the

 Wick (WL) and Gravity (GL) Lysimeters under Banana Plant

				losses of cadusafos ( $\mu$ g m <sup>-2</sup> )		percolation volumes (L m $^{-2}$ )	
decade	beginning	end	rainfall (mm)	WL	GL	WL	GL
1	10/09/01	19/09/01	44	1	1	95	15
2	20/09/01	30/09/01	105.4	6578	8778	290	40
3	01/10/01	09/10/01	193.8	6398	3786	646	138
4	10/10/01	19/10/01	203.2	200	122	666	76
5	20/10/01	31/10/01	230.4	129	27	669	78
6	01/11/01	09/11/01	6.4	10	0	31	0
7	10/11/01	19/11/01	72	49	9	192	20
8	20/11/01	30/11/01	35.8	22	2	47	1
9	01/12/01	09/12/01	122.8	47	10	360	60
10	10/12/01	19/12/01	356.2	113	32	834	134

we observed from the calculated losses by decade (**Table 2**) that the losses were maximum after application, i.e. during the second and third decades (September 20 to October 10), and accounted for 95 and 98% of total losses in the WL and GL respectively. After the third decade, losses of cadusafos were extremely small, despite percolation fluxes that remained high.

Second, we analyzed the difference between GL and WL with respect to both the concentration in percolation water and the losses of cadusafos. The closer to the application date, the greater the effect of GL on concentration and losses. Concentration was higher in GL than in WL especially during the first phase of the decay. This was brought to the fore by comparing the intercept of the two regressions shown in Figure 3: it was 794  $\mu$ g L<sup>-1</sup> for GL (95% confidence interval (CI), 468 to 1318) and 50.1  $\mu$ g L<sup>-1</sup> for WL (95% CI, 23.4 to 100). The ratio of the losses in GL to WL, i.e. the contribution of GL to the total losses, decreased from the date of application to the end of the monitoring period (Figure 4). Indeed, in spite of water fluxes being 7-fold lower in GL than in WL just after spreading during decade 2 (Table 2), the losses were of the same order of magnitude. Conversely, from the October 20 (decade 5) onward, the concentrations in WL and GL moved closer (Figure 3) and the contribution of GL to the total losses nearly equaled the contribution of GL to percolation fluxes (Figure 4).

**Cadusafos Concentration in Runoff Waters.** Concentration of cadusafos in runoff waters also followed a two-phase trend overtime (**Figure 5**), comparable to that observed in lysimeters. However, the slope of decrease of concentration in runoff was steeper than in the lysimeters: it was -0.09 (95% CI, -0.064 to 0.109), -0.12 (95% CI, -0.10- to 0.14), and -0.29 (95% CI, -0.22- to 0.36) for WL, GL, and runoff, respectively. This suggests that cadusafos became more quickly unavailable for



**Figure 5.** Concentration of cadusafos in runoff waters during the experiment. The regression was calculated for the first phase of the experiment, i.e. between September 20 and October 16.



**Figure 6.** Concentration of cadusafos in three different soil layers (0-5 cm, 5-30 cm, and 30-60 cm) under banana plants during the experiment. The vertical black arrow represents pesticide application.

runoff. The concentrations in runoff were  $1626 \ \mu g \ L^{-1}$  and  $0.2-0.4 \ \mu g \ L^{-1}$  1 day and 10 days after application, respectively. Between 10 days after application until the end of the monitoring period, the concentration of cadusafos in runoff was more steady and averaging  $0.2-0.5 \ \mu g \ L^{-1}$ , i.e. comparable to those measured in the lysimeters under the banana plants, although slightly lower at all times. There was a slight decrease in concentration though, and a regression line could be developed for this second period with a slope of -0.004917.

**Cadusafos Concentration in the Soil. Figure 6** shows that the concentration of cadusafos in soil under the banana plant was decreasing with depth and was always highest in the topsoil. No significant difference was measured between the concentration in the 5–30 cm and 30–60 cm. On September 25, on the first sampling date after application, cadusafos concentration in the topsoil was 562.8  $\mu$ g kg<sup>-1</sup>, and 66.1 and 39.5  $\mu$ g kg<sup>-1</sup> in 5–30 cm and 30–60 cm, respectively.

As with the concentration of cadusafos in runoff and drainage, we observed two phases in the decrease of concentration in soil under the banana plant with time: A steep decrease for the first 3 weeks after application (until October 17), where it reached 49.8  $\mu$ g kg<sup>-1</sup> soil in the top layer, and an order of magnitude less in the 5–30 and 30–60 cm: 5.6 and 4.0  $\mu$ g kg<sup>-1</sup> soil, respectively. Then, until the end of the monitoring period, soil concentrations were stable in the topsoil, averaging 40  $\mu$ g kg<sup>-1</sup> soil layer (from 5.6 to 8.2  $\mu$ g kg<sup>-1</sup> soil) and the 30–60 cm soil layer (between 4.0 and 7.9  $\mu$ g kg<sup>-1</sup> soil). The half-life times for cadusafos in the first phase were calculated by fitting a first-

Table 3. Total Losses of Cadusafos at the Plot Scale in Runoff and in the Wick (WL) and Gravity (GL) Lysimeters<sup>a</sup>

		losses of cadusafos (g 6000 m $^{-2}$ )		water fluxes (m <sup>3</sup> 6000 m <sup>-2</sup> )		
	surfaces (m <sup>2</sup> )	WL	GL	WL	GL	
banana plant interrow runoff total losses	611.4 5388.6 6000	8.28 (0.23%) 3.96 (0.11%) 4.74 (0.13%) 16.98	7.80 0.36	2340.6 8127 654	343.2 329.4	

<sup>a</sup> Note: the value into parentheses represents the percent of the quantity of pesticide applied initially.

order rate equation to the observed soil concentrations from the application date to October 17. The computed values were 6.6, 7.3, and 7.2 in the 0–5, 5–30, and 30–60 cm layers, respectively. These differences were not significant. Cadusafos was also measured in the soil in the interrow during the three months. The concentration remained low and averaging 13.8  $\mu$ g kg<sup>-1</sup> soil (ranging from 2.5 to 37.3  $\mu$ g kg<sup>-1</sup>), 5.8  $\mu$ g kg<sup>-1</sup> soil (ranging from 2.4 to 24.2  $\mu$ g kg<sup>-1</sup>), and 2.7  $\mu$ g kg<sup>-1</sup> soil (ranging from 1.0 to 4.6  $\mu$ g kg<sup>-1</sup>) in the 0–5, 5–30, and 30–60 cm layer, respectively. No particular temporal dynamic, however, could be distinguished from these data.

**Total Losses at the Plot Scale.** Total losses at the plot scale accounted for about 17 g on the experimental field (**Table 3**), i.e. 0.5% of the total applied amount of cadusafos. In particular, results show that 49% of the losses occurred by percolation under the banana plants, which is consistent with the high concentrations observed in percolation water in this location. The second half of losses was almost equally shared between interrow percolation (23%) and surface runoff (28%). The relative importance of the percolation losses under the interrow, despite contamination levels of percolation water being almost 1 order of magnitude smaller than those under bananas, simply arises from the large area attributed to the "between banana" surface.

#### DISCUSSION

In this paper, we addressed the problem of soil and water contamination by the nematicide cadusafos, widely used in banana cropping systems. While our results show that cadusafos losses in waters (percolation + runoff fluxes) accounted for only 0.5% of the pesticide applied initially, these losses are sufficient to contaminate water at concentrations exceeding the threshold for pesticide residue for potable water (0.1  $\mu$ g L<sup>-1</sup>). This low value was unexpected given the high water fluxes under tropical conditions. The half-life of cadusafos is particularly short, calculated to be 7 days from our field result, highlighting the high degradability of this molecule. Volatilization could be an additional dissipation pathway for cadusafos. Indeed, the molecule has a dimensionless  $K_{\rm h}$  of 5.42  $\times$  10<sup>-5</sup> which, according to the classification of Jury (35), makes it a moderately volatile pesticide while the FOOTPRINT database classifies it as "volatile" based on its saturated vapor pressure of 119.6 mPa (36). A third explanation is that despite the high rainfall volume, the runoff coefficient remained low, i.e. mostly about 10% (28), which can be explained by the saturated hydraulic conductivity for the horizon A being particularly important (135 mm  $h^{-1}$ ) (34). Finally, the low values can be explained by cadusafos retention in the soil. Acetone-extractable quantities of cadusafos do not appear particularly important (the final stock of cadusafos was about 100 g ha<sup>-1</sup> considering a final average cadusafos concentration of 70  $\mu$ g kg<sup>-1</sup>, a bulk density of 0.7, and a weight

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water content of 1000 g kg<sup>-1</sup> in the 0–5 cm soil layer), which can be a sign that bound residues are formed in large quantities, as reported by Olvera-Velona et al. (31).

Percolation fluxes mainly were involved in pesticide transport in our study: they carried 72% of the total losses while runoff carried 28%. Runoff losses are in accordance with those described in Calvet et al. (37) and Leonard et al. (8). There is little data available relating to the measurement of percolation losses of pesticides in field conditions (19) and the simultaneous measurement of losses in percolation *and* runoff (38, 39). The major transport by percolation in our study conditions indicates a particular risk for contamination of groundwaters. Although on this particular study site no water table has been detected close to the soil surface, risks of groundwater contamination are high due to the high infiltration capacity of the soil.

The main losses occurred under the banana plant where water fluxes were highest and where cadusafos was applied. Nevertheless, the losses in the interrow were unexpectedly high and reached half of those measured under the banana plant. This suggests that an important redistribution of the molecule occurs at the soil surface in the banana plantation or by subsurface lateral fluxes that can be caused by a drop in hydraulic conductivity at 30 cm below the soil surface. Such a difference in losses between the banana plant and the interrow is consistent with the result obtained in the case of fertilization (34, 40). It may occur for other crops where heterogeneous water fluxes have been observed. One implication of these results involves reconsidering the localization of cadusafos at the base of the banana plant.

Losses of cadusafos mainly originated from saturated flow as measured by GL lysimeters. The difference for drainage fluxes between WL and GL has been previously discussed by Cattan et al. (26), and we can hypothesize that the GLs collect saturated fluxes, while the WLs collect all fluxes. Several authors (41-43), referring to the dual porosity concept, suggest that zero-tension lysimeters collect essentially macropore flow whereas the wick lysimeters also collect matrix flow. Thus, the high concentrations of cadusafos in GL in comparison to the small concentrations in WL suggest that macropore flow is much more contaminated than matrix flow. This may be explained by the following. First, percolation flow is primarily contaminated when rainfall water infiltrates in the soil since the major amount of cadusafos in the soils is always at the soil surface or in the soil surface layer. Contamination arises either by dissolution of the applied cadusafos granules or by leaching of cadusafos retained in the top soil layer. As a result, the large velocity of macropore flow and the small contact it exerts with the soil matrix do not favor adsorption of the transported compounds and thereby conserves the contamination level of macropore flow whereas matrix flow, which exhibits smaller velocity and larger contact, enhances the possibilities of compound retention by the soil matrix.

Comparing the decay of cadusafos concentration in surface runoff and percolation waters gives information about the migration of cadusafos. Both decays exhibited two phases, but the duration of the phases varied slightly between surface runoff and percolation waters. This leads to three points when comparing the evolution of their contamination: (1) immediately following application, the initial cadusafos concentrations were similar in runoff and GL (for the week after the beginning of the experiment); (2) in the second week (between September 27 and October 4), cadusafos concentration decreased more rapidly in runoff than in GL which explains why the first phase lasted less for runoff water than for percolation waters; and (3) cadusafos concentrations became similar again in runoff and GL during the second phase of the experiment (starting on October 17). An explanation for point 1 is that the residues were mainly located in the first few millimeters of soil and that the compounds transported by percolation did not adsorb on the solid phase significantly enough to decrease the concentration in the percolation water collected at 60 cm depth. This is consistent with a macroporal transport previously mentioned and as described in other studies (19). For point 2 we can speculate that all the granules had progressively dissolved and the migration of cadusafos within the soil has occurred which probably depleted the very first few millimeters of the soil profile, considered to be the mixing zone between the soil and the surface runoff water (8). Consequently, the availability of cadusafos to surface runoff decreased faster than its availability to percolation. Finally, for point 3, which corresponds to the second decay phase, the very low concentrations of cadusafos in both runoff and percolation water suggest that most of the pesticide is desorbed from the soil to surface runoff and percolation to a similar extent. In this phase the observed concentrations in water were small but almost constant. They seem to represent the baseline of water contamination once the most mobile part of cadusafos due to the last application has disappeared.

Cadusafos concentration in the soil decreased in a two-phase pattern with time. The 7 days half-life calculated in the first phase indicates a fast dissipation of cadusafos that is most likely due to volatilization as discussed previously, and degradation in the soil due to conditions that are probably conducive to degradation: high temperatures and adaptation of microorganisms to cadusafos degradation due to repeated application of this molecule over a number of years, as has been shown for other pesticides (44, 45). The main databases report half-lives of 11 to 59 days (36, 46), which can be considerably shortened in soils repeatedly treated with cadusafos (47). Zhen et al. (48) investigated degradation of cadusafos in andosols and reported a half-life of 9-10 days. In the second phase, no significant dissipation occurred, which suggests that the residues stabilized in the soil. This is consistent with the phases of decrease in cadusafos concentration in surface and percolation waters during the same period, which seem clearly under the dependence of the observed cadusafos content in the soil. In the second phase, with stabilized soil contamination, the presence of cadusafos in waters can be interpreted as a slow and rate-limited desorption kinetic from the solid phase after the easily available fraction of cadusafos has been transported into waters and/or degraded.

To conclude, our study aimed at examining the transfer of cadusafos applied to banana plant bases to surface and percolation waters, in the particular context of a high spatial variability at the field scale and in tropical humid conditions. Its originality lies in studying both runoff and percolation waters. One important finding is that despite the high water fluxes under tropical conditions, only a small percentage of the applied cadusafos, (0.5%) is subject to transport in water out of the field. However, this should not be neglected since it is enough to contaminate seriously the water resources and given the number of treatments in these areas, about four per year. A second important finding is the high contribution of percolation fluxes to pesticide transport, particularly that of saturated macropore flow, which implies that the risk of groundwater contamination is large. A second implication is that reducing pollution by limiting surface runoff with grassed strips or soil treatments increasing soil infiltrability would not be appropriate in these tropical conditions because most cadusafos transfers occur by

infiltration with only small transfer via runoff. Third, our study revealed the high spatial heterogeneity of losses coming from the redistribution of rainfall by the banana canopy and from pesticide application at the base of the plant: most losses occurred under the banana plant. This heterogeneity must be accounted for in order to better assess the environmental impact of pesticide application or to establish pesticide balance at the field scale. Accordingly, the strategy of pesticide application at the base of banana plant is questioned by these results. A more even spreading should be considered under the conditions of still maintaining a proper crop protection. Finally, the study also showed that water contamination by cadusafos exhibits a twophase dynamic. In the first phase, contamination was large both in the surface and percolation waters but decreased rapidly within three weeks due to dissipation of cadusafos in the soil. In the second phase, contamination of soil and water became chronic and reached a baseline with cadusafos concentrations that were moderate but still larger than desirable with regard to drinking water standards.

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