

Pesticide Residues in Heterogeneous Plant Populations, a Model-Based Approach Applied to Nematicides in Banana (*Musa* spp.)

PHILIPPE TIXIER,^{*,†} CHRISTIAN CHABRIER,[†] AND ERIC MALÉZIEUX[‡]

CIRAD, UPR 26, PRAM - BP 214 - 97285 Lamentin Cedex 2 - Martinique, French West Indies, France, and CIRAD, UMR SYSTEM, ARGO.M, Bât. 27, 2 Place Viala, 34060 Montpellier Cedex 5, France

Nematicides are widely used to control plant-parasitic nematodes in intensive export banana (*Musa* spp.) cropping systems. Data show that the concentration of fosthiazate in banana fruits varies from zero to 0.035 g kg⁻¹, under the maximal residue limit (MRL = 0.05 mg kg⁻¹). The fosthiazate concentration in fruit is described by a Gaussian envelope curve function of the interval between pesticide application and fruit harvest (preharvest interval). The heterogeneity of phenological stages in a banana population increases over time, and thus the preharvest interval of fruits harvested after a pesticide application varies over time. A phenological model was used to simulate the long-term harvest dynamics of banana at field scale. Simulations show that the mean fosthiazate concentration in fruits varies according to nematicide application program, climate (temperature), and planting date of the banana field. This method is used to assess the percentage of harvested bunches that exceed a residue threshold and to help farmers minimize fosthiazate residues in bananas.

KEYWORDS: Nematicide residues; fosthiazate; banana; *Musa* spp.; SIMBA; population model; preharvest interval; agricultural products safety; consumer exposure to pesticides

1. INTRODUCTION

Health Hazards because of Pesticide Residues in Crops.

Pesticides are diverse and omnipresent in agriculture (1). They are toxic by nature, and hence they may cause health hazards to humans and animals through exposure or dietary intake. Human health hazards vary with the type of the pesticide and with the extent of exposure (2).

The way in which pesticides are regulated and norms are set for maximum residue levels are of importance in the context of developing country exports. Regulation authorities set an acceptable daily intake (ADI) and a maximum residue level (MRL) according to available knowledge about pesticide toxicity and according to the amount of residues found in the crop after pesticide use in the context of good agricultural practices. MRL represents the maximum concentration of a pesticide residue (expressed as mg kg⁻¹) legally permitted in food. MRLs on food imports are set by each country and are imposed as regulatory standards at the border (3, 4). Residues in crop are the result of multiple factors, including climate after the application and the dose (5). Generally, residues of pesticides on harvested products decrease with time between application and harvest, depending on the properties of the pesticides. Preharvest interval (PHI) is defined as the time between the last pesticide application and the harvest. The PHI is one of the major factors affecting pesticide residues in crops (6, 7).

Residues in Heterogeneous Plant Populations. PHI is applicable when both pesticide application and harvest occur at a precise time. Hence, a major issue is the case of heterogeneous plant populations whose products are harvested over a long period. Heterogeneous plant populations are frequently encountered in tropical contexts where plants do not follow seasonal variations as in temperate climate, for example, bananas (*Musa* spp., AAA group, cv. Cavendish Grande Naine), papaya, passion fruit, or pineapple. It is also the case for indeterminate plants, for example, strawberries, beans, or tomatoes. Some tools have to be developed to assess this food security matter and to search for solutions.

Our case study focuses on residues of fosthiazate in export bananas, the fourth highest exported food product in the world (8). In Martinique French West Indies, 29.6 tons of nematicide-active ingredients were used in 2004 for almost 8000 ha of banana fields (9). Our objective is to build a tool that helps minimize the residue in harvested fruits on the basis of a better combined management of pesticide application and harvest dates.

Banana Population from Synchronized to Unsynchronized. Export bananas currently cover nearly one million hectares worldwide (8). Bananas are rhizomatous herbs whose terminal bud produces the inflorescence. Each plant successively produces suckers issued from a lateral shoot. The sequence can be repeated for 1 to 50 generations or more, which means that it can be considered as perennial (10). The main developmental stages of banana plants include sucker appearance, growth,

* To whom correspondence should be addressed. E-mail: tixier@cirad.fr.

† CIRAD, UPR 26.

‡ CIRAD, UMR SYSTEM.

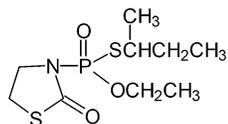


Figure 1. Structural formula of the fosfiazate (*RS*)-*S*-sec-butyl O-ethyl 2-oxo-1,3-thiazolidin-3-ylphosphonothioate (16).

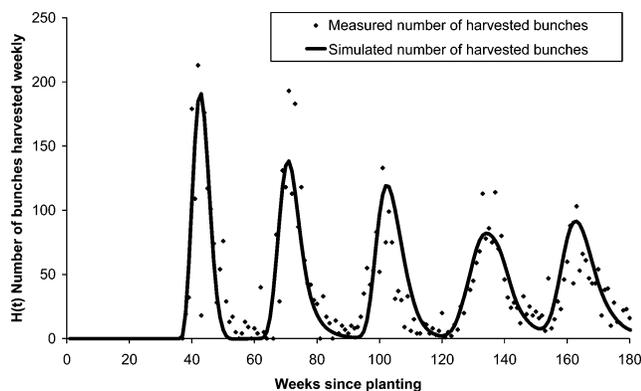


Figure 2. Harvest dynamics measured and simulated with SIMBA-POP. Measures were carried out in a field in Guadeloupe at 45 m of altitude.

flowering, and harvest. They develop at their own rhythm and do not follow a synchronous cycle. When the stages of new sucker, flowering, and harvest occur, the population structure spreads following lognormal functions (11). Hence, at any given time, a banana field consists of a population of individual plants at various developmental stages. The dynamics of banana bunch harvest follows peaks whose amplitudes tend to be wider over time, up to a continuous harvest after five to seven cropping cycles (Figure 1). The development rate of banana plants and the harvest dynamics at the field scale depend on the climate, in particular, the temperature (10, 11).

Banana Cropping Systems and Nematicide Uses. As in many intensive agrosystems, banana monocultures in the French West Indies (FWI) are hampered by major parasitic factors (plant-parasitic nematodes, insect pests, and pathogenic fungi) that seriously threaten the sustainability of these systems by decreasing yield and causing plant toppling, thus leading to intensive pesticide use. *Radopholus similis* (Cobb) Thorne and *Pratylenchus coffeae* Zimm are plant-parasitic nematodes that generate extensive root lesions, and they are considered to be among the most detrimental pathogens of banana (12). Necrosis also has harmful trophic effects such as reduced plant growth and extended vegetative period because of less efficient water and mineral uptake and transport by the roots (12–14).

In the FWI, banana growers usually apply nematicide from February to March, from May to June, and from September to October, which correspond to the most favorable climatic conditions for pesticide efficiency. The number of nematicide applications varies from one to three per year. Two applications may be considered as the more usual practice (9, 15). Nematicides include a variety of products. Among them, fosfiazate, a medium-strength systemic agent, is one of the most common nematicides used by banana producers in the FWI. It was as a result chosen for this study.

Fosfiazate. Fosfiazate is a systemic nematicide/insecticide developed by ISK Biosciences Corporation (Concord, OH) and commercialized by Syngenta (16). The IUPAC name of the fosfiazate is (*RS*)-*S*-sec-butyl O-ethyl 2-oxo-1,3-thiazolidin-3-ylphosphonothioate and its formula is shown in Figure 1. It is a systemic active ingredient used as a nematicide for banana

Table 1. SIMBA-POP Parameters^a

parameter	value
heat unit accumulated before first flowering (°C)	1750
heat unit accumulated before first sucker selection (°C)	1950
heat unit accumulated before first harvest (°C)	900
flowering lognormal stochastic curve parameter <i>a, b, c</i>	1.00, 7.00, 0.35
sucker selection lognormal stochastic curve parameter <i>a, b, c</i>	1.00, 3.00, 0.28

^a From ref (11) and not-shown experimental results.

(17, 18) and potatoes (19–22). This active ingredient belongs to the organophosphorus chemical class and inhibits acetylcholinesterase (23, 24). Nemathorin 10G is a granular formulation containing 10% active ingredient (100 g kg⁻¹). The maximal allowed application in FWI is 4 kg ha⁻¹ year⁻¹ of active ingredient. Each application consists of 15–20 grams of commercial product deposited around the banana pseudostem (usually 1850 plants per hectare). The toxic features of fosfiazate include carcinogenicity, reproductive and developmental toxicity, neurotoxicity, and acute toxicity for humans. The MRL of fosfiazate was set to 0.05 mg kg⁻¹ (23).

2. MATERIALS AND METHODS

SIMBA-POP, a Model for Simulating Plant Population Structure. The model used to simulate the harvest dynamics of banana fields is the SIMBA-POP model (11). It runs with a weekly step *t* and its structural unit is the cohort. Each cohort represents the number of plants of the same phenological stage. Two linked chains of cohorts represent the plants before flowering (preflowering cohort chain) and the plants after flowering (postflowering cohort chain). The only input data of the model is the mean weekly temperature. Outputs include *H(t)*, the number of harvested plants for each step *t*. The passage between cohorts depends on the time, the heat unit accumulated, and the stochastic laws (flowering, sucker selection, and harvest dispersions). The banana is assumed to develop only when the temperature is over 14 °C (25, 26). Therefore, the heat units accumulated per day is equal to the daily temperature minus 14. Table 1 presents the SIMBA-POP parameters used for the simulations. At each time step, SIMBA-POP simulates the banana population structure, that is, the distribution of plants according to plant and fruit development stage. As a result, and knowing the date of the last pesticide application, it is possible to calculate PHI(*t*), the preharvest interval at step *t* for every harvested plant. Figure 2 presents the measured and the simulated number of bunches harvested weekly during five cropping cycles on a field in Guadeloupe (French West Indies; 16°15 N, 61°32 W) at 45 m of altitude.

Nematicide Residues in Banana as a Function of the PHI. According to the data used in this study, we considered that fosfiazate banana fruit residue follows a Gaussian distribution function of PHI. The increasing part of the Gaussian distribution (for small PHI) traduces the delay necessary for the nematicide to be transported from soil to roots to banana fruits (approximately 3 m). The decreasing part of the Gaussian distribution (for high PHI) reflects the degradation of pesticide within the banana fruits. Such a degradation process has been recorded on other crops (5). The relationship between fosfiazate residues in banana fruits RES(*t*) and PHI(*t*) was established using data of Syngenta (18) on Nemathorin 10G. To obtain these data, 38 analyses of fosfiazate residues were performed in the pulp of bananas with 20 g of Nemathorin 10G applied per plant. The extraction was carried out by filtration and centrifugation of a banana pulp–distilled water (with 1% of NaCl) crush. This solution was passed through a 1-g C18 SPE Cartridge (Bond Elut Varian) and pesticides were released with 2.5 mL of methanol. The methanolic extract was injected in a gas chromatography column (RTX OPP Pesticide Restek), and measures were realized on a Shimadzu FPD detector. These data were used to establish the envelope Gaussian distribution of RES(*t*) in function of PHI(*t*) (Figure 3). The PHI was comprised between 7 and 100 days.

Calculation of the Concentration of Fosfiazate in the Harvested Bunches. To calculate the potential concentration of fosfiazate in

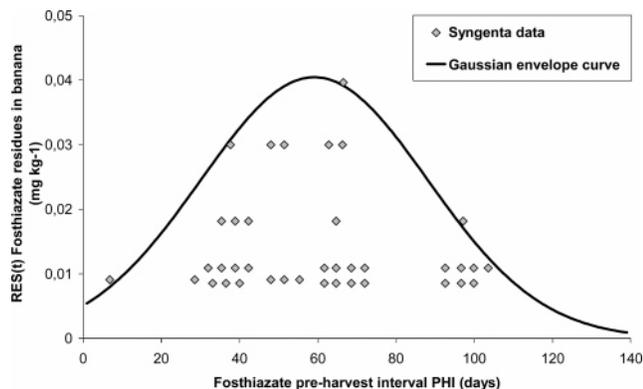


Figure 3. Residues of fosthiazate in banana fruit (mg kg^{-1}) for different nematicide applications, harvest interval (in days) data from Syngenta (18), fitted envelope Gaussian curves.

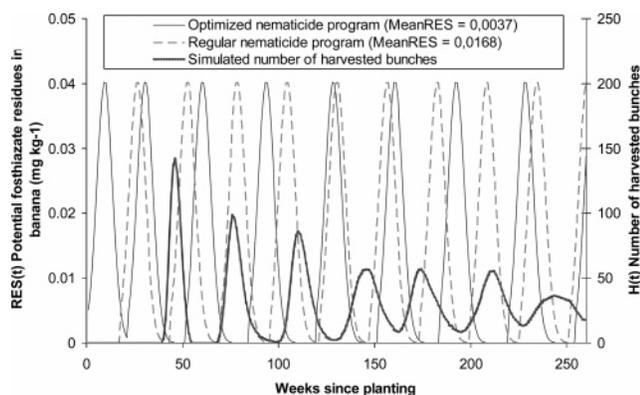


Figure 4. Simulation of nematicide residues in banana fruit for a regular and for an optimized nematicide program. Simulations were done for conditions at 350 m of altitude, with two nematicides per year (week 18 (+52i) and 44 (+52i) for the regular program and weeks 1, 22, 52, 85, 120, 152, 184, 220, and 255 for the optimized program).

banana fruits at step t of the simulation, we used the envelope curve of residues in banana pulp (Figure 3). This approach assesses the maximal risk of nematicide residues. Only the effect of the last application was taken into account because the interval between two applications is always longer than 4 months. Equation 1 presents the Gaussian curve that serves to calculate at step t the potential nematicide residue concentration $\text{RES}(t)$ in function of the nematicide PHI at step t $\text{PHI}(t)$.

$$\text{RES}(t) = a \exp\left[-\frac{1}{2} \left(\frac{\text{PHI}(t) - b}{c}\right)^2\right] \quad (1)$$

The three parameters a , b , and c of the Gaussian curve are 0.04, 59.12, and 29.00, respectively.

We consider that at each time step the concentration of fosthiazate in harvested banana fruits is equal to $\text{RES}(t)$. The next step of the approach consisted of weighting $\text{RES}(t)$ with the number of harvested bunches at step t . MeanRES, the mean concentration of fosthiazate in banana fruit over the whole simulation (n time steps), is calculated in eq 2. We made the assumption that the weight of bunches is constant over the simulation.

$$\text{meanRES} = \frac{\sum_{t=1}^{t=n} (H(t) \text{RES}(t))}{\sum_{t=1}^{t=n} H(t)} \quad (2)$$

3. RESULTS

Figure 4 presents the simulated potential residue concentration over the simulated period in banana fruit for two nematicide application programs. The regular program corresponds to a systematic application of fosthiazate at weeks 18 (middle of

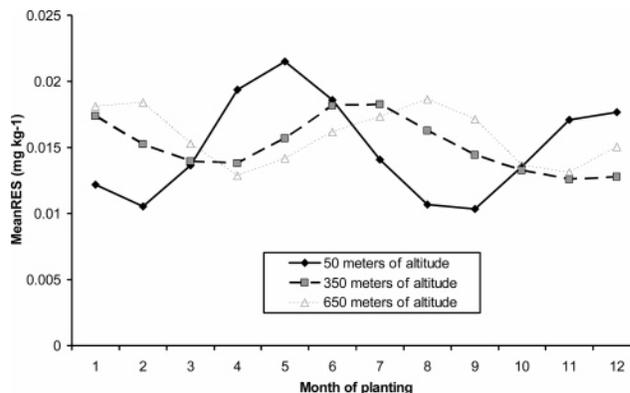


Figure 5. Mean concentration of fosthiazate in banana fruit over a 5-year simulation (with a regular nematicide program: weeks 18 and 44 every year) for every possible month of planting and for three altitudes (with a mean climatic year).

dry season) and 44 (end of rainy season) every year. The optimized program has the same number of nematicide applications over the simulation but they are chosen to minimize $\text{RES}(t)$. The date of each nematicide application was chosen to occur between two harvesting peaks. The two different nematicide application programs lead to different potential residue concentrations at a given date (Figure 4). As a result, it is possible to reduce meanRES significantly (0.0168 – $0.0037 \text{ mg kg}^{-1}$) by choosing the best nematicide application dates. Furthermore, Figure 3 shows that after the third harvesting peak, it is hardly possible to respect the PHI required by regulations because there are bunches harvested all year long.

We used our model to calculate the mean concentration of fosthiazate in banana meanRES over a 5-year simulation with a regular nematicide program (weeks 18 and 44 every year) for every possible month of plantation and for three altitudes with a mean climatic year (Figure 5). In the case of a regular nematicide program, there appear wide variations of potential residues when the nematicide applications occur at different times relative to harvesting peaks. At higher altitudes, the temperature decreases and the interval between two successive harvesting peaks is larger; it is therefore potentially easier to design nematicide programs that minimize the residues because of the bigger gap between two harvesting peaks. This highlights the importance of determining the dates of nematicide applications and the predominance of the date of nematicide application over the climatic variations and the date of planting.

We used our model (for a 5-year simulation at 350 m of altitude with a regular nematicide program: weeks 18 and 44 every year) to measure the number of bunches that exceed a nematicide residue threshold (Figure 6). Every threshold value and possible months of planting are tested. It shows that if bunches that exceed half of the maximum residues allowed ($\text{MRL}/2 = 0.025 \text{ mg kg}^{-1}$) are eliminated, 29% of the harvest is lost. This appears not to be compatible with the farm constraints. In spite of this result, it is necessary to consider that this study deals with potential residues and thus has to be considered as a risk management tool for farmers.

4. DISCUSSION

Limit of the Methodology. This modeling approach is based on the dynamics of banana populations (Figure 2) and on the residue curve that is function of the PHI (Figure 3). The main limit of the methodology lies in the residue curve. Indeed, the evaluation depends on its shape, amplitude, and magnitude. We

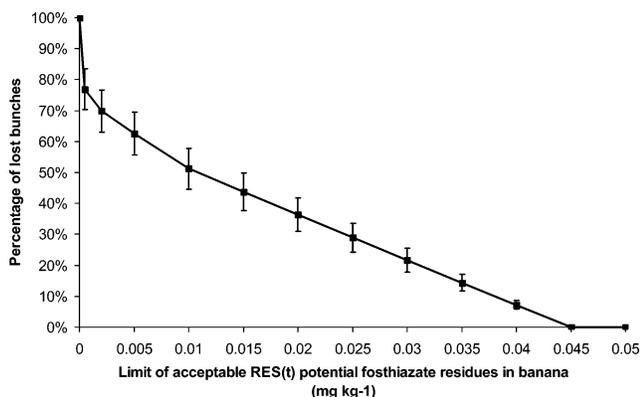


Figure 6. Mean value and standard deviation for the percentage of bunches lost in function of the acceptable potential fosthiazate residues in banana. Simulations done for 350 m of altitude and for all possible months of planting.

propose here to consider the envelope curve, which permits one to assess the potential risk, the first goal of our methodology. It probably overestimates the residues in fruits in most cases, but in the case of risk management, the worst case has to be considered. Another factor that may modify the residue content is the degradation during the transport between the area of harvest and the area of consumption. However, for given areas of production and consumption, the harvest–consumption interval is relatively constant. Finally, the condition of growth of the fruits, including the climatic conditions or the soil type, may also influence the residue content. Hence, in future studies this point should be studied more specifically.

After describing and parameterizing the conceptual framework that allows tackling the pesticide residues in heterogeneous plant populations, the next step will be to carry on a validation of the model predictions by measuring pesticide residues in bananas at harvest.

This method is complementary to some modeling and probabilistic approaches for assessing consumer exposure to pesticides (27, 28). The high variability of RES for a similar PHI may be explained in part by the variation of soil and climate conditions in which the measures have been carried out. The degradation of fosthiazate in soil depends on the soil type (29, 30), and thus it may be more relevant to investigate the effect of soil type on a fruit's residues.

The Respect of Regulations. In this paper, we highlight a major issue about the application of pesticides on heterogeneous plant populations. We show it may be difficult to respect regulations for the PHI. In our case study, on the basis of data used for calibration, the risk to have residues that exceed the MRL in bananas is nil. This tool is in line with agricultural good practices and serves in the definition of safer pest control practices (31). It is also noticeable that to not exceed the maximal amount of active ingredient applied to the field allowed per year, farmers may adjust the nematicide quantity to the number of plants (taking into account the plants that died) and also use other active ingredients. In our study, we considered simple nematicide programs with only the fosthiazate; in further research, we need to consider nematicide programs with two or more active ingredients.

Toxicology, Environmental Risk, and Nematicide Efficiency. If well managed, the first three cropping cycles of banana may lead to no residues in fruits. The environmental impact of pesticides and their efficacy on the nematode populations are other issues of concern. It could be of interest

to link the residue-minimizing methodology we propose here with other modeling approaches such as (1) environmental impact indicators like the Rpest indicator that dynamically evaluates the pollution risk by pesticides (32) or (2) the SIMBA-NEM model (33) that forecasts the nematode population dynamics and the effect of nematicides on their dynamics. These linked models should be useful to search for the best trade-offs between minimizing residues and environmental impacts and maximizing nematicide efficacy. These prospects highlight the need for a comprehensive approach in designing new cropping systems.

LITERATURE CITED

- (1) Tilman, D.; Cassman, K. G.; Matson, P. A.; Naylor, R.; Polasky, S. Agricultural sustainability and intensive production practices. *Nature* **2002**, *418*, 671–677.
- (2) Hamilton, D.; Ambrus, Á.; Dieterle, R.; Felsot, A.; Harris, C.; Petersen, B.; Racke, K.; Wong, S.-S.; Gonzalez, R.; Tanaka, K.; Earl, M.; Roberts, G.; Bhula, R. Pesticide residues in food - acute dietary exposure. *Pest Manage. Sci.* **2004**, *60*, 311–339.
- (3) European Union. Regulation (EC) No. 396/2005 of the European Parliament and of the Council of 23 February 2005 on maximum residue levels of pesticides in or on food and feed of plant and animal origin and amending Council Directive 91/414/EEC; 2005.
- (4) Wilson, J.; Otsuki, T. To spray or not to spray: pesticides, banana exports, and food safety. *Food Policy* **2004**, *29*, 131–146.
- (5) Zhang, Z.-Y.; Liu, Y.-J.; Yu, X.-Y.; Zhang, C.-Z.; Hong, X.-Y. Pesticide residues in the spring cabbage (*Brassica oleracea* L. var. *capitata*) grown in open field. *Food Control* **2007**, *18*, 723–730.
- (6) Cengiz, M.; Certel, M.; Karakas, B.; Göçmen, H. Residue contents of captan and procymidone applied on tomatoes grown in greenhouses and their reduction by duration of a pre-harvest interval and post-harvest culinary applications. *Food Chem.* **2007**, *4*, 1611–1619.
- (7) Gambacorta, G.; Faccia, M.; Lamacchia, C.; Di Luccia, A.; La Notte, E. Pesticide residues in tomato grown in openfield. *Food Control* **2005**, *16*, 629–632.
- (8) FAO, Food and Agriculture Organization Statistical Databases. <http://faostat.fao.org/faostat/> (accessed Dec 2005).
- (9) Chabrier, C.; Mauléon, H.; Bertrand, P.; Lassoudière, A.; Quénéhervé, P.; Banane Antillaise. les systèmes de culture évoluent. En Martinique, méthodes alternatives pour réduire l'utilisation des nématocides et insecticides en bananeraie. *Phytoma* **2005**, *584*, 12–16.
- (10) Turner, D. W. Banana and plantain. In *Environmental Physiology of Fruit Crop*; CRC Press, 1994; Vol. 2, pp 37–64.
- (11) Tixier, P.; Malézieux, E.; Dorel, M. SIMBA-POP: a cohort population model for long-term simulation of banana crop harvest. *Ecol. Model.* **2004**, *180*, 407–417.
- (12) Gowen, S.; Quénéhervé, P. Nematode parasites of bananas, plantains and acaba. In *Plant parasitic nematodes in subtropical agriculture*; Luc, M., Sikora, A., Bridge, J., Eds.; CAB International, 1990; pp 431–460.
- (13) Seinhorst, J. W. Water consumption of plants attacked by nematodes and mechanisms of growth reduction. *Nematologica* **1981**, *27*, 34–51.
- (14) Fogain, R. Effect of *Radopholus similis* on plant growth and yield of plantain (*Musa*, AAB). *Nematology* **2000**, *2*, 129–133.
- (15) Henriques, W.; Jeffers, R. D.; Lacher, T. E., Jr.; Kendall, R. J. Agrochemical used on banana plantation in Latin America: perspectives on ecological risk. *Environ. Toxicol. Chem.* **1997**, *16*, 91–99.
- (16) Tomlin, C. D. S. *The pesticide manual*, 13th ed.; British crop protection council Editor: Farnham, Surrey, U.K., 2004; pp 501–502.

- (17) Chabrier, C.; Hubervic, J.; Quénéhervé, P. Evaluation of fosthiazate (Nemathorin (R) 10G) for the control of the nematodes in banana fields in Martinique. *Nematropica* **2002**, *32*, 137–147.
- (18) Syngenta. *Nemathorin, le nématicide et l'insecticide de la banane*. Technical facts sheet, 2002.
- (19) Grove, G.; Woods, S. R.; Haydock, P. P. Toxicity of 1,3-dichloropropene and fosthiazate to wireworms (*Agriotes* spp.). *Ann. Appl. Biol.* **2000**, *137*, 001–006.
- (20) Back, M. A.; Haydock, P. P. J.; Jenkinson, P. The effect of granular nematicides on the development of *Rhizoctonia solani* diseases and their interaction with *Globodera rostochiensis* on potato. *BCB Conf. Pest Dis.* **2002**, *1*–2,777–780.
- (21) Syngenta. *Nemathorin 10G, insecticide du sol contre les nématodes et les taupins*; Technical facts sheet, 2004.
- (22) Minnis, S. T.; Haydock, P. P. J.; Evans, K. Control of potato cyst nematodes and economic benefits of application of 2,3-dichloropropene and granular nematicides. *Ann. Appl. Biol.* **2004**, *145*, 145–156.
- (23) EPA. *Fosthiazate fact sheet*; U.S. Environmental Protection Agency, Office of Prevention, Pesticides, and Toxic Substances, U.S. Government Printing Office: Washington, DC, 2004.
- (24) Hurley, M.; Balboa, A.; Lushington, G.; Guoc, J. Interactions of organophosphorus and related compounds with cholinesterases, a theoretical study. *Chem-Biol. Interact.* **2005**, 157–158, 321–325.
- (25) Ganry, J.; Meyer, J. P. Recherche d'une loi d'action de la température sur la croissance des fruits du bananier (Research of a temperature law on the growth of the fruits of banana). *Fruits* **1975**, *30*, 375–392.
- (26) Turner, D. W.; Lahav, E. The growth of banana plants in relation with temperature. *Aust. J. Plant Physiol.* **1983**, *10*, 43–53.
- (27) Hamey, P.; Harris, C. The variation of pesticide residues in fruits and vegetables and the associated assessment of risk. *Regul. Toxicol. Pharm.* **1999**, *30*, S34–S41.
- (28) Caldas, E. D.; Boon, P. E.; Tressou, J. Probabilistic assessment of the cumulative acute exposure to organophosphorus and carbamate insecticides in the Brazilian diet. *Toxicology* **2006**, *222*, 132–142.
- (29) Qui, S.; Gan, J.; Liu, W.; Becker, O. J. Degradation and Adsorption of Fosthiazate in Soil. *J. Agric. Food Chem.* **2004**, *52*, 6239–6242.
- (30) Pantelelis, I.; Karpouzas, D. G.; Menkissoglu-Spiroudi, U.; Tsiropoulos, N. Influence of Soil Physicochemical and Biological Properties on the Degradation and Adsorption of the Nematicide Fosthiazate. *J. Agric. Food Chem.* **2006**, *54*, 6783–6789.
- (31) Gomes da Cruz, A.; Cencib, S. A.; Maia, M. C. A. Good agricultural practices in a Brazilian produce plant. *Food Control* **2006**, *17*, 781–788.
- (32) Tixier, P.; Malézieux, E.; Dorel, M.; Bockstaller, C.; Girardin, P. Rpest a dynamic indicator to assess pesticide water pollution risk. Application to banana-based cropping systems in FWI. *Eur. J. Agron.* **2007**, *26*, 71–81.
- (33) Tixier, P.; Malézieux, E.; Risède, J. -M.; Dorel, M. Modelling populations of banana phytoparasitic nematodes: a contribution to the design of sustainable cropping systems. *Ecol. Model.* **2006**, *198*, 321–331.

Received for review September 21, 2006. Revised manuscript received January 12, 2007. Accepted January 15, 2007.

JF062710F