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Field-Incurred Fenitrothion Residues in Kakis: Comparison of Individual Fruits, Composite Samples, and Peeled and Cooked Fruits

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Field trials have been carried out to determine the variability of residue levels of fenitrothion and its main metabolites fenitrothion-oxon and 3-methyl-4-nitrophenol in individual kaki fruits versus composite samples, in peel versus flesh, and in whole uncooked versus whole cooked fruits. Residue levels have been determined by gas chromatography with thermionic specific detection after extraction with ethyl acetate and without further cleanup. At harvest, residue levels of fenitrothion were below maximum residue levels (MRLs) and the two metabolites 3-methyl-4-nitrophenol and fenitrothion-oxon could be quantified with average amounts of 0.080 and 0.012 mg/kg, respectively. Levels of fenitrothion decreased 88% after peeling, whereas temperature did not result in a high variation. The ratios of the highest residue level in the individual fruits to the corresponding mean of residue levels in the composite samples for fenitrothion were <3. This value is lower than that recommended by the World Health Organization as default value for consumer risk assessment.

KEYWORDS: Residue; fenitrothion; metabolites; variability factor; processing; *Diospyros kaki*; minor crop

INTRODUCTION

Kaki (*Diospyros kaki*) is a deciduous fruit tree adapted to warm temperate and subtropical climates. It is originally from China and Japan. The major producers are China, Japan, Brazil, Korea, and Italy. Minor producers include Israel, the United States, New Zealand, Australia, Spain, Egypt, India, and Chile (*1*). The income-producing regions in Spain of this minor crop are Andalucía and Valencia; 70% of the production is exported mainly to the European market (*2*). Kaki fruits have been found to be an excellent source of vitamin A, and antioxidative properties as β-carotenes have been identified (*3*).

The nonsystemic organophosphate (OP) insecticide fenitrothion (*O*,*O*-dimethyl *O*-4-nitro-*m*-tolyl phosphorothioate) is oxidized by mono-oxygenases in animals, insects, and plants to the metabolite fenitrothion-oxon (**Figure 1**), which is a more powerful inhibitor of cholinesterase than the parent (*4*, *5*). The major excretion product is the metabolite 3-methyl-4-nitrophenol (**Figure 1**), the potential mutagenic effect of which is not clear (*4*, *6*−*8*). In Spain, fenitrothion is registered in kaki for control of cochineal insects, aphids, and San Jose scale (*Quadraspidiotus perniciosus*). The recommended preharvest interval (PHI) is 15 days. The European maximum residue level (MRL) for fenitrothion in the group of miscellaneous fruits is 0.5 mg/kg.

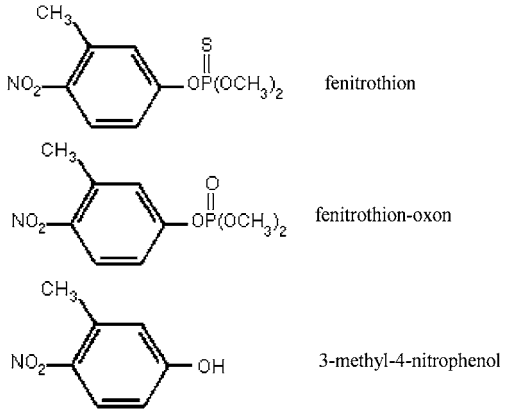

Figure 1. Chemical structures of fenitrothion, fenitrothion-oxon, and 3-methyl-4-nitrophenol.

Acute and long-term dietary risk assessment is estimated by considering the amount of a residue (parent compound and relevant metabolites) in or on food and the amount of food consumed. Data on residue levels on treated raw commodities are necessary for the establishment of MRLs permitted in that commodity as are data on the fate of these residues during food processes such as peeling or cooking for the estimation of the real residue intake (*9*−*12*). MRLs are based on composite samples, in which a number of individual commodity items are homogenized and the analysis is carried out on the mixed

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sample. With recent attention being focused on the potential for acute exposure through diet, the possible risk associated with the presence of higher residues on individual food items than in the composite samples has been considered (*9*−*12*). For consumer risk assessment, default values for variability factors (v) (ratio of the highest level of residue in the individual size samples to the corresponding residue levels in the composite samples) have been set at 5 for large crops (commodity weights > 250 g) and at 10 for medium crops (commodity weights between 25 and 250 g) (*9*).

The aims of this study were to determine residue levels of fenitrothion and its main metabolites in kaki fruits after field treatment and to study the variation of these residue levels in individual kaki fruits versus composite samples, in peel versus flesh, and in whole uncooked versus whole cooked fruits.

Although several analytical methods based on gas chromatography have been developed for the quantification of fenitrothion in different plant materials (*13*−*15*), few attempts to detect the metabolites fenitrotion-oxon and 3-methyl-4-nitrophenol in foods have been reported. An analytical method for the simultaneous determination of these compounds in the peel, flesh, whole fruit, and boiled fruit has been developed and validated in our laboratory by gas chromatography with thermionic specific detection (TSD) after extraction with ethyl acetate, a frequently used solvent for monitoring of organophosphate pesticide residues in vegetables (*16*, *17*).

MATERIALS AND METHODS

Field Trials. Three different sites located in eastern Spain (Comunidad Valenciana) were selected for the study. In each site two plots were chosen as control plot and treated plot with four to nine trees in each. The trees were 7, 9, and 12 years of age in each site, respectively. Their size was approximately 3 m high by 3.5 m diameter, and the planting distance was 5 × 4.5 m. The application rate was the higher authorized in Spain for fenitrothion in kaki trees. Fenitrotion 50% (EC) w/v was sprayed as recommended with a manual sprayer at 0.15% (75 mL in 50 L of water). A single application of fenitrothion was made on October 7, 8, and 15, 2002, to each site, respectively. An agrometeorological station located near each plot collected meteorological data. During the whole experiments, the minimum/maximum daily air temperatures and total rainfall registered in the three sites were between 7 and 9 °C and between 31 and 34 °C, and between 78 and 81 mm, respectively.

Samplings. Samples were gathered before application of the product (0), 3 h postapplication, and 15 days postapplication (recommended preharvest interval, PHI). At each time, ∼24 kaki fruits weighing ∼4 kg were collected randomly per plot, taking fruits from all parts of four trees (top, bottom, inside, and outside) as recommended by European guidelines (*18*) and transported at 4 °C and in darkness in labeled polyethylene bags to the laboratory, where samples were processed and stored at −20 °C until analysis. The fruits of each plot were subsampled and subjected to the following studies at each time: (a) six fruits were analyzed individually as whole raw kakis and (b) six fruits were peeled with a kitchen knife. The thickness of the peel removed in that way was ∼2 mm. Composite samples of flesh and pulp were analyzed separately. (c) Six fruits were homogenized for residue level analysis of the raw composite, and 500 g was boiled without water during 15 min for analysis. Weights of individual kaki fruits ranged between 160 and 300 g.

Analytical Procedure. *Reagents.* Cyclohexane and ethyl acetate were of HPLC grade from Lab-Scan Ltd. (Dublin, Ireland). Sodium sulfate anhydrous, granular for analysis, was purchased from Merck (Darmstadt, Germany). Individual stock standard solutions were prepared in a solution of cyclohexane/ethyl acetate (9.5:0.5) from analytical grade material of purity = 98.5% for fenitrothion, 96.7% for fenitrothion-oxon, and 99.0% for 3-methyl-4-nitrophenol, obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Concentrations ranged from 1 to 1000 ng/μL. Mixed standard solutions were prepared from the stock solutions at concentrations of 0.01, 0.05, 0.1, 0.5, 1, and 2 μg/mL. Helium, nitrogen, and air were from Air Liquide España (Madrid, Spain).

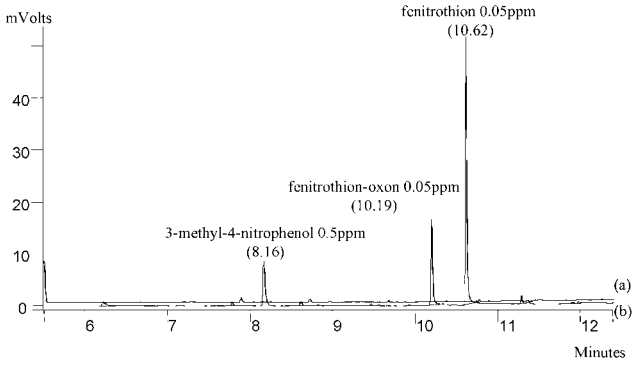

Figure 2. Chromatograms of (**a**) kaki blank and (**b**) mixed standard solution in cyclohexane/ethyl acetate (9.5:0.5) of 3-methyl-4-nitrophenol (0.5 μg/mL), fenitrothion-oxon (0.05 μg/mL), and fenitrothion (0.05 μg/mL).

Extraction. Twenty-five grams of whole raw kaki, pulp, and cooked kaki homogenized in a blender was mixed in a rotary stirrer (5 min) with two portions of 50 mL of ethyl acetate and 20 g of sodium sulfate. For the analysis of peel, 2.5 g of finely cut kaki was blended with two portions of 25 mL of ethyl acetate and 2 g of sodium sulfate. When applicable, spiking solutions were added to the matrix for recovery trials. The homogenate was centrifuged in a Sorvall RC5C Plus centrifuge at 4000 rpm (2430*g*) and ambient temperature during 15 min. After shaking and centrifugation, the organic phases collected from the two extractions were pooled and dried in a Heidolph rotary evaporator model Laborota 4000 at 50 °C. The residue was dissolved in 20 mL of a solution of cyclohexane/ethyl acetate (9.5:0.5); 2.0 μL of this solution was injected in the gas chromatograph system.

GC Determination. For compound determination, a Varian CP-3800 gas chromatograph equipped with a thermionic specific detector and a split−splitless injector model 1079 (Varian Iberia, S.L., Madrid, Spain) was used. Sample extracts were injected into the GC/TSD system under the following conditions: splitless injection mode; injection volume, 2.0 μL; injector temperature, 300 °C; detector temperature, 300 °C; and carrier gas, helium. The gas chromatograph was operated in constant flow mode at 1 mL/min. GC temperature program: hold for 2.0 min at 70 °C, increase to 200 °C at 30 °C/min, hold for 1 min, increase to 220 °C at 10 °C/min, hold for 0 min, and finally increase to 300 °C at 30 °C/min and hold for 10 min. A Chrompack capillary column CP-Sil 8 CB low bleed/MS, 30 m × 0.25 mm i.d. and a 2-m length of fused silica tubing uncoated 0.25 mm i.d. purchased from Varian Iberia (Madrid, Spain) were used.

RESULTS AND DISCUSSION

Analytical Method. With the described method fenitrothion, fenitrothion-oxon, and 3-methyl-4-nitrophenol could be determined with sufficient sensitivity and reproducibility. The concentrations of compounds in the sample extracts were obtained by comparing peak areas from samples with those recorded for the standards. Retention times of the compounds were 8.1, 10.1, and 10.6 min for 3-methyl-4-nitrophenol, fenitrothion-oxon, and fenitrothion, respectively (**Figure 2**). No interfering peaks appeared at the retention times of the compounds in the blank whole kaki and pulp chromatograms, but in the 25 g blank peel and cooked sample an interfering peak with the metabolite 3-methyl-4-nitrophenol was identified at concentrations <0.02 mg/kg. Fortified samples at two concentration levels were run, and the quantification was carried out using calibration curves of six calibration points of mixed standard solutions at 0.01, 0.05, 0.1, 0.5, 1, and 2 μg/mL made up in cyclohexane/ethyl acetate (9.5:0.5). The linearity was very good (R^2 > 0.99). Repeatability at 0.05 and 0.5 μg/mL of five

Table 1. Data of the Analytical Method

compound	fortification level (μg/mL)	recovery (%) (RSD, %)	
3-methyl-4-nitrophenol	0.05	whole kaki, pulp	80 (13.4)[a]
		peel	78 (14.9)[a]
		boiled kaki	
	0.50	whole kaki, pulp	80 (4.2)[a]
		peel	73 (4.4)[a]
		boiled kaki	93 (13.9)[a]
fenitrothion-oxon	0.01	whole kaki, pulp	100 (14.2)[b]
		peel	78 (7.0)[b]
		boiled kaki	102 (7.9)[b]
	0.10	whole kaki, pulp	108 (9.8)[b]
		peel	99 (8.2)[b]
		boiled kaki	111 (11.4)[b]
fenitrothion	0.05	whole kaki, pulp	98 (10.1)[a]
		peel	80 (15.9)[a]
		boiled kaki	78 (8.4)[a]
	0.50	whole kaki, pulp	112 (4.4)[a]
		peel	92 (5.5)[a]
		boiled kaki	100 (13.0)[a]

[a] Mean of five replicates. [b] Mean of three replicates

Table 2. Residues [Mean ± RSD (%), $n = 3$] of Fenitrothion and Its Main Metabolites in Composite and Processed Samples of Kaki Fruits

time[a]	3-methyl-4-nitrophenol (mg/kg)	fenitrothion-oxon (mg/kg)	fenitrothion (mg/kg)
	Whole Kaki		
3 h	nd[b]	<0.008	0.68 ± 9
15 days	0.080 ± 57	0.012 ± 20	0.22 ± 31
	Flesh		
3 h	<0.040	<0.008	0.014 ± 54
15 days	0.053 ± 35	<0.008	0.018 ± 39
	Peel		
3 h	<0.040	<0.008	6.10 ± 17
15 days	<0.040	<0.008	0.99 ± 27
	Boiled Whole Kaki		
3 h	<0.040	<0.008	0.56 ± 6
15 days	0.093 ± 49	0.010 ± 19	0.19 ± 16

[a] Time after application. [b] Not detected.

replicates gave good relative standard deviation (RSD < 15%). Satisfactory recoveries ranging between 73 and 112% were obtained as shown in **Table 1**. Quantification limits (LOQ) were 0.008 mg/kg for fenitrothion-oxon and fenitrothion in whole kaki, pulp, peel, and boiled fruit, 0.04 mg/kg for 3-methyl-4-nitrophenol in whole kaki and pulp, and 0.08 mg/kg in peel and boiled kaki.

Kaki Residues. The residues of fenitrothion, fenitrothion-oxon, and 3-methyl-4-nitrophenol in composite samples of whole raw kaki fruits from each site are reported in **Table 2**. Fenitrothion and its main metabolites could be identified in analytical samples 3 h and 15 days (proposed PHI) post-treatment. No interfering peaks were observed in control plots and in samples harvested before treatment. Residue levels of fenitrothion declined from 0.68 to 0.22 mg/kg, 3 h to 15 days post-treatment, respectively. At harvest, these residue levels are below the established European MRLs of 0.5 mg/kg for fenitrothion in kaki fruits. Immediately after application, trace amounts of the metabolite fenitrothion-oxon could be detected. At PHI, the two metabolites could be quantified in samples with average amounts of 0.080 and 0.012 mg/kg for 3-methyl-4-nitrophenol and fenitrothion-oxon, respectively. The relevance of these metabolites for consumer risk assessment should be taken into account as they are present at concentrations >0.01 mg/kg (*19*) and as toxicological properties for these metabolites have been reported in the literature (*4*−*8*).

Table 3. Residues of Fenitrothion and Its Main Metabolites Fenitrothion-oxon and 3-Methyl-4-nitrophenol in Individual Size and Composite Samples of Kaki Fruits

time after application	sample	3-methyl-4-nitrophenol (mg/kg)	fenitrothion-oxon (mg/kg)	fenitrothion (mg/kg)
3 h	individual kaki fruits ($n = 18$)			
	mean ± RSD (%)	0.078 ± 54[a]	0.012 ± 16[b]	0.73 ± 45
	min/max values	nd[f]/0.17	nd/0.014	0.12/1.42
	composite samples ($n = 3$)			
	mean ± RSD (%)	nd	<0.008	0.68 ± 9
	min/max values	nd	<0.008/0.0084	0.62/0.74
	variability factor[e]			2.1
15 days	individual kaki fruits ($n = 18$)			
	mean ± RSD (%)	0.11 ± 62[c]	0.016 ± 43[d]	0.21 ± 62
	min/max values	nd/0.31	nd/0.033	0.042/0.54
	composite samples ($n = 3$)			
	mean ± RSD (%)	0.080 ± 57	0.012 ± 20	0.22 ± 31
	min/max values	0.041/0.13	0.010/0.015	0.15/0.29
	variability factor[e]	3.9	2.7	2.4

[a,b,c,d] Mean and RSD have been calculated for 10, 7, 14, and 13 fruits, respectively; values for the other fruits were <LOQ. [e] Ratio of the highest residue level in the individual fruits to the corresponding mean of residue levels in the composite samples. [f] Not detected.

Effect of Household Processing on Residue Levels. Kaki fruits are eaten normally without their peel, and they can be used in pastry. We have studied the distribution of residue levels of fenitrothion and its metabolites in pulp and peel and the effect of temperature on levels of these compounds.

Fenitrothion is a nonsystemic insecticide. Results from peel and flesh reported in **Table 2** indicate that fenitrothion residues practically remain in peel, as only small amounts were identified in the flesh, which can be in part the result of contamination during the process of peeling. The percentage weights of pulp and flesh in kaki fruits are 11 and 89%, respectively. The distribution of fenitrothion between peel and flesh 15 days after treatment is 88%. This result is consistent with those reported in previous studies in carrots with the nonsystemic organophosphate insecticides diazinon and parathion and the nonsystemic pyrethroid insecticide cypermethrin, where residues present in whole carrot were not detected after peeling (*20*). Similar results were observed with the nonsystemic phenylcarbamate insecticide chlorpropham in potatoes, where the amount of residue removed by peeling was >90% (*21*). It seems that fenitrotion-oxon and 3-methyl-4-nitrophenol enter into the flesh more easily than their parent compound as trace amounts were detected in the peel and flesh 15 days after treatment, and the levels observed in the flesh were of the same order as that found in whole kaki.

Temperature did not result in an evident increase or decrease of residue levels of fenitrothion and the two main metabolites after boiling without water during 15 min (**Table 2**).

Residue Variability among Individual Fruits. The residues of fenitrothion and its main metabolites in individual fruits versus composite samples are reported in **Table 3**. The residues of fenitrothion in individual fruits ranged from 0.12 to 1.42 mg/kg, 3 h post-treatment, and from 0.042 to 0.54 mg/kg, 15 days post-treatment. The residue concentrations in composite samples of whole raw kakis taken 3 h and 15 days postapplication ranged from 0.62 to 0.74 mg/kg with an average of 0.68 mg/kg and an RSD of 9% and from 0.15 to 0.29 mg/kg with an average of

0.22 mg/kg and an RSD of 31%, respectively (**Table 3**). From these results variability factors (ν) of 2.1 and 2.4 were estimated for 3 h and 15 days postapplication, respectively.

A comparison of the concentration levels of fenitrothion, 15 days after treatment, in the individual fruits with the average concentrations in the composite samples shows that only three individual fruits contained higher residues than the average value (0.22 mg/kg), that is, 0.40, 0.43, and 0.54 mg/kg. The residue levels of fenitrothion in the 18 fruits were lower than the European MRL except for one fruit having 0.54 mg/kg.

Residues of 3-methyl-4-nitrophenol in individual fruits ranged between not detected and 0.17 mg/kg, 3 h post-treatment, and from not detected to 0.31 mg/kg 15 days post-treatment (**Table 3**). No residues of this metabolite were detected 3 h post-treatment in the composite sample. The residue concentrations of this metabolite in composite samples of whole raw kakis taken 15 days postapplication ranged from 0.041 to 0.13 mg/kg, with an average of 0.080 mg/kg and an RSD of 57% (**Table 3**). From these results a variability factor (ν) of 3.9 is estimated as the ratio of the highest residue level found in individual fruits to the corresponding residue level seen in the composite sample.

The residues of fenitrothion-oxon in individual fruits ranged from not detected to 0.014 mg/kg, 3 h post-treatment, and from not detected to 0.033 mg/kg, 15 days post-treatment (**Table 3**). Trace amounts of the metabolite in composite samples of whole raw kakis were detected 3 h after application, and 15 day postapplication residue levels ranged from 0.010 to 0.015 mg/kg with an average of 0.012 mg/kg and an RSD of 20% (**Table 3**). From these results a variability factor (ν) of 2.7 was estimated 15 days post-treatment.

A variability factor of up to 2.5 can be set for risk assessment of fenitrothion in kaki fruits. This value is lower than the variability factors recommended by the World Health Organization to be used as default values (*9*). Studies in oranges with aldicarb (*22*) and in potatoes with clorpropham (*21*) also showed that variability factors estimated were lower than the recommended default factors for consumer risk assessment.

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