

# MULTIRESIDUE ANALYSIS OF OVER 233 PESTICIDES IN CUCUMBER AND GRAPEFRUIT SAMPLES USING A QuEChERS AND GAS CHROMATOGRAPHY-TANDEM MASS SPECTROMETRY-BASED METHOD

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**Abstract.** This report describes the 233 pesticide residue in cucumber and grapefruit sold in Turkey. In total, 680 fruit and vegetable samples were analyzed by liquid chromatography connected to tandem mass spectrometry (LC-MS / MS) with the device in which gas chromatography to mass spectrometry (GC-MS) units work together. In cucumber, limit of detection (LOD) varied from 0.17  $\mu\text{g kg}^{-1}$  to 11.8  $\mu\text{g kg}^{-1}$  and the limit of quantifications (LOQ) varied from 0.42  $\mu\text{g kg}^{-1}$  to 39.35  $\mu\text{g kg}^{-1}$ ; whereas for grapefruit, LOD, and LOQ values varied from 0.13  $\mu\text{g kg}^{-1}$  to 11.80  $\mu\text{g kg}^{-1}$ , 0.42  $\mu\text{g kg}^{-1}$ , and 39.35  $\mu\text{g kg}^{-1}$ , respectively. Recoveries obtained for each pesticide in both matrices ranged between 77.87% and 104.15%. Relative standard deviation (RSD) values calculated were  $> 20\%$  (24.32%) for clofentazine in cucumber at 20.0  $\text{mg kg}^{-1}$  and (21.36%) for propyzamide, (29.11%) for fludioxonil grapefruit at 100  $\mu\text{g kg}^{-1}$ . For the other compounds, at both concentration levels evaluated in cucumber and grapefruit, the RSD values were  $< 20\%$ . The extended measurement uncertainty for individual pesticides ranged from 0.208 to 0.434. In general, 11.6% of cucumbers and 13.8% of grapefruits contained at least one detectable residue, but levels were below the EU legal limits.

**Keywords:** *QuEChERS, GS-MS, LC-MS / MS, pesticide, grapefruit, cucumber*

## Introduction

Pesticide is used in all kinds of chemical substances and preparations used in agricultural researches and applications. The pesticide may be a chemical agent, a biological agent such as a virus or a bacterium, an antimicrobial, a disinfectant or any other vehicle. Although the use of pesticides may have some benefits, it can cause problems for humans and other living things due to their potential toxicity. Such as pesticides, some of which include biopreparations, insect and plant growth regulators, pheromones (hormone-bearing) and other attractants, feed-blockers, repellents (insect repellents), traps, plant activators, preparations used in the treatment of physiological diseases (Epstein and Zhang, 2014).

Turkey is the 4th largest fresh fruit and vegetable producers in the world. As of 2015, about 37% of the total production of the citrus group consisting of lemon, orange, mandarin and grapefruit (3.7 million tons) and cucumber (100 thousand tons) accounted for approximately 2.8% of exports. Almost all of the total value of exported fresh fruits and vegetables has entered the EU, Russian Federation, Ukraine and Iraqi markets (TUIK, 2015; FAO, 2015).

In food, the permissible residual limit values are as low as in parts per million (ppm) and in parts per billion (ppb). Therefore, it is accepted that the most effective approach in pesticide residue analysis is multiresidue analyzes. In recent years, many different methods have been successfully used in the detection of pesticide residues in

foods, especially fruits and vegetables. In order to use these methods gas chromatography (GC), liquid chromatography (LC), coupled to mass selective detectors (MS), many new sample preparation techniques and extraction methods have been developed. Many sample preparation techniques for the detection of many pesticides used in vegetables and fruits, including solid-phase extraction (SPE) (Campone et al., 2018; Chan et al., 2018), solid-phase microextraction (SPME) (Li et al., 2019; Dong et al., 2019), accelerated solvent extraction (ASE) (Su et al., 2018; Stachniuk, 2018), supercritical fluid extraction (SFE) (Alvarez et al., 2019; Khan et al., 2018), matrix solid-phase dispersion (MSPD) (Liu et al., 2018; Jiang et al., 2018), microwave-assisted extraction (MAE) (Du et al., 2018; Wang et al., 2018), membrane extraction (Salemi et al., 2019), gel permeation chromatography (GPC) (Zhu et al., 2019), and QuEChERS (quick, easy, cheap, effective, rugged and safe) method (Tankiewicz, 2019), have been used. Within these techniques, QuEChERS has become more preferred in recent years, because it reduces the use of solvents in accordance with the 'green chemistry' principles, and provides a simple, inexpensive, fast and reliable result.

In this study, 205 pesticides of different classes, which are suitable for LC-MS/MS analysis, and 28 pesticides for GC-MS analysis, were discussed. In our country, pesticide use is still indispensable within the framework of integrated pest management principles. Selected pesticides, widely used in the production stages of fruits and vegetables in Turkey. Some of the selected pesticides may cause acute and chronic diseases due to their toxicity (Kumar et al., 2018).

In Turkey, along with concepts such as food security, have been a lot of studies in a very food production due to the increased use of pesticides. For example, tomatoes (Cengiz et al., 2018), baby foods (Kilic et al., 2018), oranges (Golge and Kabak, 2015), fish products (Polat et al., 2018), apricots (Ersoy et al., 2018), olive oil (Razzaghi et al., 2018) and milk (Yıldırım et al., 2018). However, there are very little data about the presence and levels of pesticide residues in cucumber and grapefruit consumed in Turkey.

This report aims to determine multi-residue pesticides in cucumber and grapefruits consumed in Turkey, monitor the compliance of the fruit and vegetables with the legal limits for target pesticide residues. In this context, the validated analytical method based on gas chromatography to mass spectrometry (GS-MS) and liquid chromatography coupled with tandem mass spectrometry (LC-MS/MS) was applied to 680 samples for a total of 233 pesticides for the periods 2015–2017.

## **Material and Method**

### ***Sample collection***

A total of 680 fruit and vegetable samples, including 330 cucumber and grapefruits 350 samples, were obtained from three provinces of Turkey, namely Hatay, İskenderun, Adana, Mersin and Antalya. Approximately 2 kg of cucumber and grapefruit samples were supplied from bazaar, supermarkets (2015–2017). The vegetables were transported quickly to the laboratory, chopped and homogenised with a home food processor (Robot Coupe model R10, Turkey). The chopped samples were then stored at  $-18^{\circ}\text{C}$  until sample preparation step.

### ***Reagents and chemicals***

Standards of pesticides detected in GC-MS were supplied from ChemService (West Chester, PA, USA) and Standards of pesticides detected in LC-MS/MS from Sigma – Aldrich (St. Louis, MO, USA). All standards were of  $\geq 98\%$  purity. Stock solution of multistandard mixture at a concentration of  $10 \text{ mg L}^{-1}$  for each substance was dissolved in acetonitrile and kept at  $-18^\circ\text{C}$  in the dark. This solution was used for preparation of matrix-matched calibration standards at concentrations varying from  $0.1$  to  $100 \text{ }\mu\text{g kg}^{-1}$  and for spiking the QuEChERS extracts in validation study.

Acetone, acetonitrile and methanol (GS-MS and LC-MS/MS grade), Isooctane, cyclohexane, ethyl acetate, magnesium sulphate, sodium acetate, glacial acetic acid, formic acid and ammonium formate (purity  $\geq 99.9\%$ ) were supplied from Sigma – Aldrich (Stenheim, Germany). Helium (purity  $\geq 99.9\%$ ) was supplied by Praxair (Colima, Mexico). Ultrapure water of  $18.2 \text{ M}\Omega$  was purchased using a Milli Q water purification system (Milford, MA, USA). The primary-secondary amine (PSA) was supplied from Supelco (Darmstadt, Germany).

### ***Sample preparation***

Pesticide residues were extracted from cucumber and grapefruit samples using QuEChERS protocol according to AOAC official method (AOAC, 2007) with some changes. Cucumber representing high water content product groups, grapefruit with high acid and high water content groups were selected as samples for this study. These vegetable and fruit were selected because of their economic importance in export. Fifteen grams of homogenised sample was placed in to a 50 mL polypropylene tube, then 15 mL methanol-acetic acid (99:1, v/v), 6 g  $\text{MgSO}_4$  and 1.5 g sodium acetate were added. The mixture was shaken vigorously by vortex mixer (Vortex Jr. Mixer from American Scientific Products, catalog no. S82251-1) for 1 min and centrifuged (Eppendorf 022628225 5804R Centrifuge with A-2-DWP-Plate, Germany) for 2 min at 2000 rpm. Later, 8 mL of the upper methanol phase was added in a 15 mL polypropylene tube containing 900 mg of  $\text{MgSO}_4$  and 150 mg of PSA. The mixture was vortexed for 1 min, followed by centrifugation for 2 min at 2000 rpm. For each sample, 200  $\mu\text{L}$  was injected into GS-MS and LC-MS / MS.

For GC-MS, 0.010, 0.025, 0.05, 0.1, 0.2, 0.5 and 1 ppm prepared standards were given to the device and the corresponding area and retention time were determined. Calibration curve is obtained by plotting the area graph against concentration. For LC-MS/MS, 0.005, 0.010, 0.020, 0.040, 0.080 and 0.160 ppm prepared standards were delivered to the device and the area corresponding to these concentrations and retention time were determined. The fortified samples were released to equilibrate for 80 minutes before treatment. Blank samples were also prepared according this procedure.

### ***GC-MS conditions***

Analyses were carried out with a Agilent GC-MS, 5975C inert MSD Series gas chromatograph equipped with an MS detector. The system was equipped with capillary column (Rtx-440 GC). Oven start temperature was set at  $110^\circ\text{C}$ , increased to  $250^\circ\text{C}$  at a rate of  $35^\circ\text{C}/\text{min}$ , increased again at a rate of  $40^\circ\text{C}/\text{min}$  to  $280^\circ\text{C}$ , this temperature was kept for 3 minutes. The total run time was 5.5 minutes. One mL of sample was injected in split mode (2:1). The injector temperature was set at  $250^\circ\text{C}$ .

The helium carrier gas flow was 1.1 mL/min. The mass spectrometer was operated in electron impact ionization mode at 70 eV of collision energy with 2 minute solvent delay to prevent damage to the filament of the ion source. The temperature of the ion source, manifold, and transfer line was set at 250°C, 40°C, and 280°C, respectively. For qualitative analysis a full scan from 75 mass/charge (m/z) to 280 mass/charge (m/z) was applied. For quantitative pesticide analysis preferred ion monitoring (SIM) mode was used.

### ***LC-MSMS conditions***

Analyses were carried out with a ThermoQuest Surveyor MS Pump and autosampler. Chromatographic separation was carried out on a Waters C18 analytical column Phenomenex LUNA 5 µm C18 150 x 2 mm from Phenomenex (Torrance, CA, USA). The system was controlled by Xcalibur Home Page, Version 1.2 software. The column temperature was set at 40°C. The solvent systems used were (A) 0.1% acetic acid and 10 mM ammonium acetate in water and (B) 0.1% acetic acid and 10 mM ammonium acetate in 95:5 CH<sub>3</sub>CN:Water. A flow rate of 200 µL min<sup>-1</sup> and an injection volume of 10 µL were applied.

The MS was operated using a Negative Ion Electrospray – with metal needle option. The metal needle was connected with a zero dead volume union to the fused silica capillary delivering the mobile phase to the ESI source. Device parameters optimized for MS are as follows. Precursor ion (m/z): 321, product ions (m/z): 257, 194, 176, 152, spray voltage: 1.5 kV, collision voltage: 26 V, source offset voltage: 5 V, electron multiplier voltage: 1.27 kV, capillary temperature: 350 C, N<sub>2</sub> sheath gas: 80 arbitrary units, N<sub>2</sub> auxiliary gas: 35 psi, collision gas: Ar.

### ***Method validation***

The proposed method was validated following the European SANTE/11945/2015 Guideline (European Commission, 2015). The established GC-MS and LCMS-MS method were validated in terms of its linearity, limit of detection (LOD), limit of quantification (LOQ), selectivity and recovery. Linearity was evaluated by constructing calibration curves with standard solutions.

### ***Limit of detection (LOD), limit of quantification (LOQ)***

For detection limit of detection (LOD) and limit of quantification (LOQ) operation; In GC-MS, 20 µg kg<sup>-1</sup> kg and LC-MS/MS 10 µg kg<sup>-1</sup> recovery was performed. 205 pesticides were injected 10 times with the LC-MS/MS device and 28 pesticide GC-MS devices. The LOD values and LOQ values were calculated according to the following formula: LOD = 3s, LOQ = 10s, s: Standard deviation.

### ***Linear measurement range***

In order to determine the linear measurement range; Standards for 10 different concentrations (5, 10, 20, 40, 80, 160 µg kg<sup>-1</sup>) for LC-MS / MS, 7 different concentrations for GC-MS (10, 25, 50, 100, 200, 500, 1000 µg kg<sup>-1</sup>) prepared, injected into devices and calibration graphs are drawn.

## Precision

### Repeatability

For repeatability study; A total of 40 recovery studies were performed by 4 analysts for GC-MS at a concentration of 20  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$ , for LC-MS/MS at concentrations of 10  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$ , each with 5 replicates.

For repeatability control, RSD% values were calculated for each person and for each concentration separately. For each individual and for each concentration, the RSD values were calculated from the mean ( $\bar{x}$ ) and standard deviation (s) of the measured concentrations, separately. Then the RSD<sub>pool</sub> values were calculated and the repeatability control was performed:  $\text{RSD} = \frac{s}{\bar{x}} \cdot 100$

### Reproducibility

Intra-laboratory reproducibility study; 20  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$  for GC-MS, LC-MS/MS at concentrations of 10  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$ , by 4 analysts for each at five different times, recovery work, was carried out. For each concentration, RSD<sub>WR</sub> values were calculated and reproducibility checked.

### Accuracy

Reality study for accuracy was done with recovery. For the reality parameter; A total of 40 recovery studies were carried out by 4 analysts at concentrations of 20  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$  for GC-MS 10  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$  for LC-MS/MS each. For reality control, all recovery values must be in the range of 70-120%.

Percentage recovery (%R) values were calculated for each individual, each concentration and each repetition.  $\%R = \frac{(C2-C1)}{C3} \cdot 100$ , C1: Measured sample concentration (blank), C2: Measured, spike made sample concentration (blank+spike), C3: Spike made concentration.

### Uncertainty assessment

The standard uncertainty ( $u_x$ ) from reality is equal to the standard deviation ( $S_{\bar{x}}$ ) of the mean, if the unit average recovery value ( $\bar{R}$ ) is not significantly different from 1. t test deđer was applied to determine if the unit mean recovery value was significantly different from 1.

- If  $t < t_{\text{table}}$ , then the reality standard uncertainty ( $u_x$ ) is equal to the, mean deviation ( $S_{\bar{x}}$ ) of the mean.
- If  $t \geq t_{\text{table}}$ , the elevated standard uncertainty ( $u_i$ ) value should be calculated.

$$u_i = \sqrt{(|1 - \bar{R}|/k)^2 + (u_x)^2} \quad (\text{Eq.1})$$

where

k: Scope factor (k = 2, for 95% confidence interval)

$\bar{R}$ : Unit average recovery value.

$$\bar{R} = \frac{\%R}{100} \quad (\text{Eq.2})$$

$$S_{\bar{x}} = \frac{s}{\sqrt{n}} \quad (\text{Eq.3})$$

$$t = \frac{|1 - \bar{R}|}{S_{\bar{x}}} \quad (\text{Eq.4})$$

where

$S_{\bar{x}}$ : Standard deviation of mean

s: Standard deviation

n: Total concentrations and total number of repetitions for all contacts.

In cucumber and grapefruit, the standard uncertainty from reality and the relative standard uncertainty were calculated using the *Equations 1, 2, 3 and 4*.

$$RSD_{pool} = \sqrt{\frac{(n_1-1) \times RSD_1^2 + (n_2-1) \times RSD_2^2 + \dots + (n_n-1) \times RSD_n^2}{(n_1-1) + (n_2-1) + \dots + (n_n-1)}} \quad (\text{Eq.5})$$

In cucumber and grapefruit, relative standard uncertainty from repeatability was calculated using *Equation 5*.

$$RSD_{WR(pool)} = \sqrt{\frac{(n_1-1) \times RSD_{WR1}^2 + (n_2-1) \times RSD_{WR2}^2}{(n_1-1) + (n_2-1)}} \quad (\text{Eq.6})$$

In cucumber and grapefruit, standard uncertainty from reproducibility was calculated using *Equation 6*. Relative standard uncertainty values are used in the total combined uncertainty calculation; however, since the relative standard deviation values are already used in the reproducibility calculations as in the reproducibility uncertainty calculations, the calculated standard uncertainty value is also equal to the relative standard uncertainty value.

## Results and discussion

### *Method validation and application*

Selectivity was evaluated by checking the presence of coextracted interferences in the chromatograms from blank samples. Hence, chromatograms of blank and fortified (100 µg/kg) samples of cucumber and grapefruit were compared (*Figure 1*). The presence of matrix interferences were interpreted by monitoring the SIM chromatograms for each pesticide at the retention time window expected for each compound. It can be observed that there are no extracted matrix interferences for the determination of the pesticides studied in both fruits; therefore this method can be considered as selective.

The matrix compatible standard calibration curves showed smooth linearity over a concentration range of 20- 100 µg kg<sup>-1</sup>. As shown in *Figure 2*, correlation coefficients were > 0.98. The values of Limits of detection (LOD) ranged from 0.17 to 11.8 µg kg<sup>-1</sup> and Limits of quantification (LOQ) ranged from 0.42 to 39.35 µg kg<sup>-1</sup> in cucumber, whereas for grapefruit, LOD, and LOQ values varied from 0.13 µg kg<sup>-1</sup> to 11.80 µg kg<sup>-1</sup>, 0.42 µg kg<sup>-1</sup>, and 39.47 µg kg<sup>-1</sup>, respectively.

The baseline (basal signal) in blank samples at the time window in which each pesticide is expected is different due to the quantification being made using different

ions. Therefore, the LOD and LOQ calculation are different for each compound. With these results it can be concluded that the method has good sensibility for cucumber and grapefruit fruits because LOD and LOQ values of all pesticides are  $<0.50$  mg / kg. The LOQ were well below the maximum residue levels (MRL) established by European Union for regulated pesticide residues both in cucumber and grapefruit, except for methylparathion, malathion and diazinon.

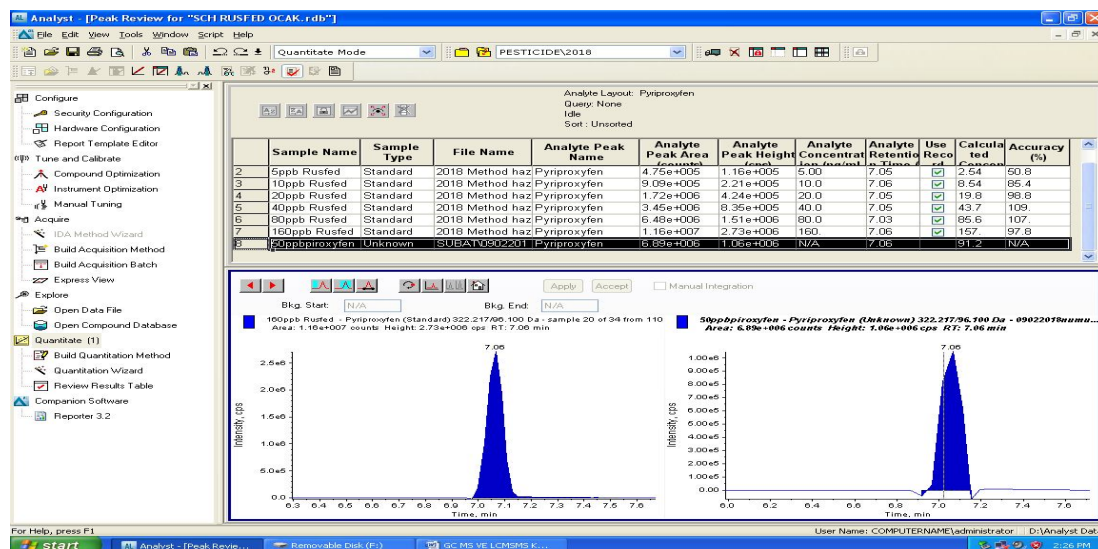


Figure 1. Chromatograms (SIM mode) obtained for pyriproxyfen injection of blank and fortified samples of cucumber and grapefruit for checking matrix interferences. Pesticide concentration was  $100\mu\text{g/L}$

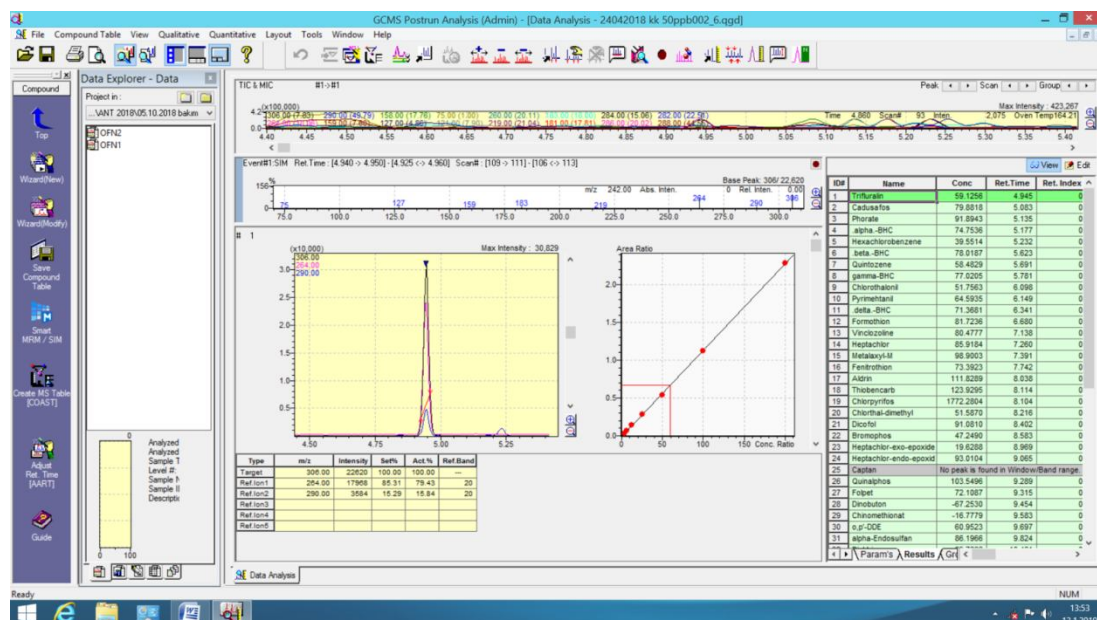


Figure 2. Chromatograms (SIM mode) obtained for trifluralin injection of blank and fortified samples of cucumber and grapefruit for checking matrix interferences. Pesticide concentration was  $100\mu\text{g/L}$

The recovery values correspond to the lowest and highest concentration of the linear range. The recovery results are as shown in *Table 1*. Recoveries obtained for each pesticide in both matrices ranged between 77.87% and 104.15%. Thus, the recovery data is in the range of values required by the SANTE Guidelines, which recommends a recovery rate of 70-120% (European Commission, 2015).

Precision was expressed as relative standard deviation (RSD) and evaluated in different days (see *Table 1*). For this purpose, 20  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$  for GC-MS, LC-MS / MS at concentrations of 10  $\mu\text{g kg}^{-1}$  and 100  $\mu\text{g kg}^{-1}$ , by four analysts for each at five different times, recovery work, was carried out. Relative standard deviation (RSD) values calculated were > 20% (24.32%) for clofentazine in cucumber at 20.0  $\text{mg kg}^{-1}$  and (21.36%) for propyzamide, (29.11%) for fludioxonil grapefruit at 100  $\mu\text{g kg}^{-1}$ . For the other compounds, at both concentration levels evaluated in cucumber and grapefruit, the RSD values were < 20%. The RSD values obtained are considered satisfactory according to the European SANTE Guideline (European Commission, 2015).

### ***Pesticide analysis in commercial fruit and vegetable samples***

The validated analytical method was used to analyse 233 pesticide residues in cucumber and grapefruit samples collected from bazaar, supermarkets in four provinces of Turkey. In our monitoring program, a total of 680 vegetable samples, including 75 cucumbers and 110 grapefruits in 2015, 90 cucumbers and 120 grapefruits in 2016, and 165 cucumbers and 120 grapefruits in 2017, were analysed. Of the 680 fruit and vegetable samples, 11.6% (35 samples) of cucumbers and 13.8% (39 samples) of grapefruits contained at least one detectable residue, but levels were below the EU MRL. There were 1 acaricide, 3 insecticides and 5 fungicides in cucumbers, while 4 insecticides and 4 fungicides were detected in grapefruit samples.

While 27 cucumber samples gave positive results, multiple pesticide residues were found in 8 samples, corresponding to 2.4% of the total samples. Clofentezine was the most frequently detected pesticide measured in cucumber with a percentage occurrence value of 4.4%, and in concentrations ranging from 0.021 to 0.042  $\text{mg kg}^{-1}$ . This insecticide is an acaricide used in various products (fruit crops, leafy vegetables, some fruiting vegetables, citrus fruits, pome fruits, grapes, cotton etc.) against red spider species. Acaricides in this group are contact effective. They also have a gastric venom effect (European Commission, 2008; Van Leeuwen et al., 2010).

The fungicide propyzamide was the second most abundant pesticide (199 out of 330 samples) in cucumbers, with levels ranging from 0.002 to 0.009  $\text{mg kg}^{-1}$ . Propyzamide, effectively controls a wide range of herbicides in several agricultural crops, including hard-shelled fruits, in grape fruits, and tree nuts. It has a chronic toxic effect (European Commission, 2008; Tian et al., 2018).

The fungicide propamocarb was the third most abundant pesticide (230 out of 330 samples) in cucumbers, with levels ranging from 0.022 to 0.032  $\text{mg kg}^{-1}$ . Propamocarb, which belongs to the class of carbamate, is used in cucumber against mildew. Does not have toxic effects (FAO, 2012; Bhat et al., 2018).

Azinphosethyl was detected individually in 6 cucumber samples (1.8%) in concentrations between 0.013 and 0.016  $\text{mg kg}^{-1}$ . It has a protective and therapeutic properties. Alternaria, Botrytis, Pythium, Rhizoctonia, Venturia and many other diseases are widely used as a fungicide. It has no acute and chronic effects (Polyiem et al., 2018).

**Table 1.** LOD, LOQ study results for GC-MS and LC-MS / MS

Pesticides	Cucumber						Grapefruit					
	LOD	LOQ	Precision		Recovery		LOD	LOQ	Precision		Recovery	
			RSD (%)		(%)				RSD (%)		(%)	
			20	100	20	100			10	100	10	100
(µg/kg)						(µg/kg)						
Trifluralin	8.08	26.94	7.45	8.65	101.91	88.39	1.04	19.26	9.15	12.54	101.91	87.45
Chlorothalonil	4.78	15.94	8.65	7.23	100.96	85.72	2.03	17.28	9.12	14.56	94.87	100.69
Formothion	6.37	21.25	9.21	6.14	96.5	88.88	2.96	17.14	10.12	17.85	95.67	91.24
Vinclozolin	5.76	19.2	11.45	8.74	104.15	91.82	2.38	10.89	15.14	19.63	104.15	96.78
Fenitrothion	8.27	27.58	10.54	9.65	100.6	88.4	4.99	5.98	14.12	15.36	100.6	85.92
Dicofol	5.66	18.87	9.65	7.32	103.01	92.15	3.85	10.66	11.1	19.36	103.01	88.41
Captan	7.31	24.38	13.65	7.65	102.61	83.91	4.53	10.14	13.1	18.36	102.61	90.69
Folpet	6.8	22.67	18.65	6.84	93.95	88.28	3.65	8.52	10.21	10.15	95.62	87.3
Qinomethionate	10.15	33.83	11.32	6.45	99.51	91.2	3.97	5.03	10.14	17.32	93.73	103.21
Dinobuton	11.8	39.35	9.32	6.21	98.75	89.45	5.26	15.42	9.85	18.36	94.54	92.38
Endosulfan A	7.56	25.2	8.65	7.52	99.77	88.52	3.64	6.89	7.65	11.1	99.77	99.56
Endosulfan B	7.38	24.61	7.45	6.36	101.08	89.17	2.99	3.48	12.69	11.2	101.08	91.18
Bromopropylate	7.49	24.96	14.12	15.36	102.61	78.55	4.77	6.77	11.36	10.21	102.61	88.69
Tetradifon	6.88	22.93	15.36	19.36	102.27	89.25	2.57	9.86	10.87	9.65	102.27	93.87
Alfa-Cypermethrin a	7.38	24.58	18.69	11.36	102.82	90.64	3.76	7.93	10.96	8.65	102.82	86.17
Alfa-Cypermethrin b	7.2	24.02	14.65	10.15	97.51	91.77	4.71	16.63	10.23	9.67	92.82	99.31
alpha.-BHC	3.96	13.19	15.21	17.32	98.26	91.28	3.02	12.82	10.65	8.63	98.99	97.52
delta.-BHC	5.94	19.79	17.45	18.36	99.6	88	2.46	15.11	11.23	8.65	99.6	90.54
Heptachlor	7.2	23.99	18.21	11.1	101.72	80.22	4.83	12.16	12.32	9.45	101.72	90.64
2,4-DDE	4.45	14.85	19.32	11.2	101.45	83.73	5.13	13.23	11.56	14.65	101.45	100.53
Dieldrin	4.24	14.13	18.65	10.54	98.88	83.67	5.54	17.53	11.45	19.32	98.88	90.56
4,4-DDE	5.78	19.26	15.32	14.32	101.79	94.44	3.76	12.14	10.65	18.65	101.79	89.22
2,4-DDD	4.8	16.01	11.35	12.36	99.8	88.56	4.32	9.97	9.63	15.32	99.8	93.71
Endrin	3.49	11.64	9.14	10.23	102.21	93.36	6.01	15.89	7.23	11.35	102.21	99.19
Chlorfenapylyl	5.82	19.4	8.54	17.36	100.93	90.78	4.44	8.56	9.63	9.14	100.93	93.53
4,4-DDD	4.84	16.12	11.21	14.65	102.11	95.72	4.88	12.54	10.25	8.54	102.11	99.48
Ethion	8.6	28.67	12.41	13.85	101.54	87.48	3.07	15.69	14.52	11.21	101.54	91.5
DDT	6.69	22.31	16.74	14.87	102.37	91.95	4.75	10.05	11.52	12.41	102.37	98.37
Abamectin	4.07	13.56	18.69	14.23	87.45	89.17	4.12	8.21	10.65	16.74	88.39	92.89
Acetamiprid	4.32	14.41	17.21	16.65	100.69	91.6	2.87	16.09	10.65	18.69	85.72	90.59
Acetochlor	4.2	14.01	11.32	18.98	91.24	88.53	2.42	17.09	11.23	17.21	88.88	91.85
Acrinathrin	4.75	15.82	8.14	9.47	96.78	89.55	0.49	18.45	11.25	14.32	91.82	90.79
Alachlor	5.04	16.79	7.65	8.31	85.92	89.36	3.01	12.54	10.87	12.63	88.4	91.43
Amitraz	3.23	10.75	10.14	7.54	88.41	86.21	5.85	14.41	9.63	14.36	92.15	90.31
Atrazine	2.43	8.1	14.65	7.98	90.69	93.05	3.26	20.03	8.35	12.31	83.91	94.33
Azinphos methyl	5.01	16.68	14.12	7.47	87.3	89.89	2.4	14.81	8.65	11.32	88.28	90.22
Azoxystrobin	6.17	20.58	11.25	6.32	103.21	89.93	4.97	16.27	7.65	10.63	91.2	104.09
Bensulfuron-Methyl	0.13	0.42	17.32	14.36	92.38	95.37	4.14	10.24	8.32	9.36	89.45	94.94
Bentazone	3.09	10.3	18.36	12.31	99.56	90.67	4.42	15.84	9.63	9.56	88.52	91.4
Beta-Cyfluthrin	3.39	11.3	11.1	11.32	91.18	91.11	3.51	13.73	12.54	8.36	89.17	89.6

Pesticides	Cucumber						Grapefruit					
	LOD	LOQ	Precision		Recovery		LOD	LOQ	Precision		Recovery	
			RSD (%)		(%)				RSD (%)		(%)	
			20	100	20	100			10	100	10	100
(µg/kg)						(µg/kg)						
Bifenthrin	2.09	6.96	11.2	10.63	88.69	86.67	5.55	9.57	14.56	9.1	78.55	98.32
Boscalid	4.23	14.1	10.21	9.36	93.87	88.36	5.94	8.08	17.85	9.89	89.25	92.43
Bromoxynil	2.68	8.92	9.65	9.56	86.17	87.84	3.3	1.63	19.63	8.63	90.64	87.48
Bromuconazole	4.18	13.92	8.65	8.36	99.31	90.22	4.69	10.02	15.36	9.52	91.77	92.72
Bupirimate	5.81	19.37	9.67	9.1	97.52	87.81	3.6	19.5	19.36	9.38	91.28	87.53
Buprofezine	3.56	11.87	8.63	9.89	90.54	88.94	4.66	10.86	11.36	10.54	88	89.81
Carbaryl	1.8	5.99	8.65	14.65	90.64	88	5.16	8.45	18.65	11.52	80.22	86.85
Carbendazim	1.24	4.12	9.45	12.32	100.53	93.49	4.08	16.56	11.32	7.65	83.73	83.68
Carbofuran	2.33	7.76	14.65	17.14	90.56	90.44	4.13	13.8	9.32	12.69	83.67	94.06
Carbosulfan	3.65	12.16	17.23	11.65	89.22	87.56	5.98	14.75	8.65	11.36	94.44	91.79
Carboxin	2.43	8.11	15.21	12.31	93.71	84.98	1.34	11.69	7.45	10.87	88.56	91.78
Carfentrazone-Ethyl	4.49	14.97	14.32	11.32	99.19	89.06	3.32	18.51	14.12	10.96	93.36	99.05
<b>Chlofentazine</b>	4.58	15.28	<b>24.32</b>	10.63	93.53	95.76	1.27	19.81	15.36	10.23	90.78	92.5
Chlorfluozuron	2.83	9.44	14.36	9.36	99.48	87.17	1.64	11.01	18.69	10.65	95.72	84.8
Chloridazon	4.01	13.37	12.31	9.56	91.5	96.47	4.34	15.65	14.65	11.23	87.48	95.69
Chlormequat chloride	0.65	2.17	11.32	8.36	98.37	92.06	4.99	12.14	15.21	12.32	91.95	102.17
Chlorpham	4.95	16.51	10.63	9.1	92.89	96.36	3.29	15.54	17.45	11.56	89.17	92.83
Chlorpyrifos ethyl	5.11	17.02	9.36	9.89	90.59	92.62	1.48	17.19	18.21	11.45	91.6	95.66
Chlorpyrifos Methyl	5.82	19.41	9.56	8.63	91.85	86.75	3.46	13.59	19.32	9.45	88.53	88.12
Chlorsulfuron	4.9	16.33	8.36	9.52	90.79	88.56	1.27	13.77	18.65	14.65	89.55	90.98
Clethodim	2.76	9.2	9.1	10.65	91.43	89.94	4.77	19.93	15.32	17.23	89.36	90.15
Clodinafop-Propargyl	4.97	16.57	9.89	8.36	90.31	86.66	1.28	4.47	11.35	15.21	86.21	93.05
Clothianidin	2.74	9.12	8.63	9.1	94.33	92.59	5.93	11.07	9.14	14.32	93.05	98.78
Cyclanilide	4.61	15.36	9.52	9.89	90.22	91.03	4.48	4.23	8.54	12.63	89.89	95.08
Cycloate	3.73	12.44	9.38	8.63	104.09	90.99	4.99	5.46	11.21	14.36	89.93	90.12
Cyhalofop-butyl	3.01	10.03	10.54	9.52	94.94	97.87	4.38	14.48	12.41	12.31	95.37	90.62
Cymoxanil	0.52	1.72	11.52	9.38	91.4	85.36	5.99	16.63	10.63	11.32	90.67	90.67
Cypermethrin	2.17	7.22	10.45	10.54	89.6	82.92	5.89	10.95	9.36	5.12	91.11	88.33
Cyproconazole	5.36	17.85	10.78	11.52	98.32	86.39	6.36	4.93	9.56	4.12	86.67	87.89
Cyprodinil	4.67	15.57	11.65	7.65	92.43	89.93	2.38	11.53	8.36	6.18	88.36	90.05
Dazomet	1.09	3.62	5.65	12.69	87.48	88.64	4.55	4.23	9.17	6.45	87.84	99.29
Deltamethrin	3.95	13.16	6.85	14.36	92.72	88.57	4.03	15.91	9.89	8.12	90.22	96.14
Diafenthiuron	3.4	11.32	7.65	12.31	87.53	86.73	4.22	4.28	8.63	9.15	87.81	94.59
Diazinon	3.46	11.53	8.41	11.32	89.81	85.88	6.06	19.77	9.52	9.12	88.94	95.27
Dicamba	3.51	11.71	8.69	10.63	86.85	88.28	2.13	14.95	9.38	10.12	88	96.27
Dichlofluanid	2.08	6.94	7.36	9.36	83.68	88.85	4.97	16.62	10.54	15.14	93.49	89.8
Dichlorvos	3.21	10.71	8.65	9.56	94.06	93.14	4.81	14.59	11.52	14.12	90.44	100.03
Diclofop Methyl	2.7	8.99	9.36	8.36	91.79	88.32	5.55	19.97	14.32	11.1	87.56	94.38
Diethofencarb	3.67	12.23	11.52	9.1	91.78	93.82	4.28	19.64	12.63	13.1	84.98	97.31
Difenoconazole	4.05	13.49	10.54	9.89	99.05	92.41	3.96	21.21	14.36	14.12	89.06	93.77
Diflubenzuron	3.53	11.78	11.57	14.65	92.5	91.96	5.7	7.92	12.31	17.52	95.76	94.41

Pesticides	Cucumber						Grapefruit					
	LOD	LOQ	Precision		Recovery		LOD	LOQ	Precision		Recovery	
			RSD (%)		%				RSD (%)		%	
			20	100	20	100			10	100	10	100
(µg/kg)						(µg/kg)						
Dimethenamid	4.68	15.6	12.65	12.32	84.8	93.18	4.37	15.17	11.32	10.32	87.17	92.01
Dimethoate	3.02	10.05	13.25	15.32	95.69	90.99	5.82	13.43	10.63	9.32	96.47	90.12
Dimethomorph	4.6	15.35	14.32	10.11	102.17	91.82	0.17	14.08	9.36	10.65	92.06	98.19
Diniconazole	4.83	16.11	14.58	11.21	92.83	83.81	5.17	20.19	9.56	11.25	96.36	86.9
Dinocap	3.06	10.2	12.69	12.32	95.66	88.55	4.21	7.12	8.36	10.87	92.62	95.16
Dioxathion	5.62	18.73	11.36	15.36	88.12	94.05	6.02	16.57	9.14	9.63	86.75	90.98
Diphenamid	3.88	12.93	10.87	19.36	90.98	92.6	5.82	16.05	9.89	8.35	88.56	100.65
Dithianon	3.19	10.63	10.96	21.36	90.15	95.55	5.75	18.48	8.63	8.65	89.94	91.63
Diuron	3.49	11.64	10.23	18.65	93.05	84.51	5.22	14.26	9.52	7.65	86.66	91.9
Epoxiconazole	6.3	20.99	10.65	11.32	98.78	91.54	4.57	13.21	9.38	8.32	92.59	96.78
EPTC	4.33	14.42	11.23	9.32	95.08	86.61	4.99	19.01	10.54	9.63	91.03	91.34
Esfenvalerate	2.96	9.86	12.32	8.65	90.12	85.69	0.55	14.56	11.52	12.54	90.99	83.28
Ethalfuralin	3.7	12.33	11.56	7.45	90.62	82.77	1.25	19.41	9.45	14.56	97.87	92.14
Ethiofencarb	1.4	4.67	11.45	14.12	90.67	84.53	2.33	0.56	8.65	17.85	85.36	90.38
Ethofumasate	4.21	14.03	10.65	9.32	88.33	89.26	4.82	17.23	7.23	9.74	82.92	99.63
Ethoprophos	4.26	14.21	9.63	10.63	87.89	88.09	2.64	14.02	6.14	10.11	86.39	93.6
Etofenprox	5.78	19.26	7.23	17.01	90.05	90.08	0.66	20.06	8.74	15.14	89.93	86.83
Etozazole	5.18	17.28	9.63	10.32	99.29	85.11	3.29	19.4	9.65	14.12	88.64	96.2
Famoxadone	5.14	17.14	10.25	11.21	96.14	93.47	5.79	19.16	7.32	13.12	88.57	92.24
Fenamidone	3.27	10.89	14.52	15.56	94.59	92.45	5.25	17.39	7.65	19.11	86.73	96.87
Fenamiphos (sum)	1.79	5.98	11.52	9.45	95.27	91.03	3.91	15.24	6.84	18.14	85.88	97.06
Fenarimol	3.2	10.66	10.65	14.65	96.27	86.62	1.69	16.63	6.45	17.14	88.28	93.21
Fenazaquin	3.04	10.14	10.65	17.23	89.8	92.99	3.55	1.82	6.21	12.11	88.85	94.41
Fenbuconazole	2.56	8.52	11.23	15.21	100.03	89.33	5.82	4.18	7.52	9.52	93.14	88.87
Fenoxaprop-P-Ethyl	1.51	5.03	11.25	14.32	94.38	78.75	4.73	7.75	6.36	8.32	88.32	89.11
Fenoxycarb	4.63	15.42	10.87	12.63	97.31	85.39	2.56	16.08	7.65	11.45	93.82	86.91
Fenpropathrin	2.07	6.89	9.63	14.36	93.77	89.51	1.57	8.79	12.69	10.65	92.41	87.7
Fenpyroximate	1.04	3.48	8.35	12.31	94.41	90.84	0.7	2.21	11.36	9.63	91.96	82.97
Fenthion	2.03	6.77	8.65	11.32	92.01	85.23	0.27	10.96	10.87	7.23	93.18	86.94
Fenvalerate	2.96	9.86	7.65	15.65	90.12	90.35	2.73	19.29	10.96	9.63	90.99	93.64
Fipronil	2.38	7.93	8.32	18.69	98.19	90.9	4.12	17.51	10.23	9.52	91.82	85.09
Fluazifop-P-Butyl	4.99	16.63	9.63	14.36	86.9	89.68	3.21	13.02	10.65	10.74	83.81	101.63
Fluazinam	3.85	12.82	12.54	7.65	95.16	85.02	3.4	5.62	11.23	11.74	88.55	92.88
<b>Fludioxynil</b>	4.53	15.11	14.56	6.84	90.98	91.03	1.75	11.82	12.32	<b>29.11</b>	94.05	93.48
Flufenoxuron	3.65	12.16	17.85	6.45	100.65	94.58	3.24	19.41	11.56	17.85	92.6	94.7
Flurasulam	3.97	13.23	19.63	6.21	91.63	92.08	3.59	15.77	11.45	19.21	95.55	92.49
Flurochloridone	5.26	17.53	15.36	7.52	91.9	83.14	0.55	8.53	8.65	8.65	84.51	90.09
Flusilazole	3.64	12.14	19.36	19.36	96.78	98.84	0.51	5.22	10.45	7.65	91.54	93.67
Flutriafol	2.99	9.97	18.36	19.36	91.34	91.02	0.79	2.35	11.74	8.32	86.61	96.84
Foramsulfuron	4.77	15.89	17.15	18.65	83.28	85.58	3.6	0.91	9.65	9.63	85.69	90.22
Formatanate	2.57	8.56	19.56	11.32	92.14	77.87	4.76	9.09	10.65	12.54	82.77	83.83
Fosthiazate	3.76	12.54	18.65	9.32	90.38	82.08	3.99	13.72	11.63	14.56	84.53	91.1

Pesticides	Cucumber						Grapefruit					
	LOD	LOQ	Precision		Recovery		LOD	LOQ	Precision		Recovery	
			RSD (%)		(%)				RSD (%)		(%)	
			20	100	20	100			10	100	10	100
(µg/kg)						(µg/kg)						
Furathiocarb	4.71	15.69	17.65	8.65	99.63	82.96	3.19	10.68	12.36	17.85	89.26	90.53
Giberellic acid	3.02	10.05	14.36	7.45	93.6	94.33	4.39	11.33	10.54	19.63	88.09	91.53
Halfenprox	2.46	8.21	17.89	14.12	86.83	95.57	8.08	5.83	11.57	15.36	90.08	100.83
Haloxyfop-2-Etoxyethyl	4.83	16.09	17.25	15.36	96.2	86.59	4.78	10.8	12.65	19.36	85.11	91.07
Haloxyfop-P-Methyl	5.13	17.09	14.56	14.36	92.24	87.72	6.37	11.98	13.25	12.36	93.47	90.46
Hexaconazole	5.54	18.45	14.85	21.36	96.87	89.58	5.76	1.84	14.32	10.15	92.45	82.71
Hexaflumuron	3.76	12.54	12.65	11.32	97.06	87.05	8.27	1.7	14.58	15.36	91.03	90.08
Hexythiazox	4.32	14.41	15.23	11.25	93.21	91.54	5.66	2.64	10.32	14.36	86.62	90.99
Imazalil	6.01	20.03	12.14	14.32	94.41	87.12	7.31	11.99	10.74	11.36	92.99	91.16
Imazamox	4.44	14.81	18.23	12.36	88.87	86.71	6.8	15.87	11.35	11.32	89.33	87.18
Imazapic	4.88	16.27	15.65	11.17	89.11	89.99	10.15	13.31	12.65	11.25	78.75	88.97
Imazapyr	3.07	10.24	18.56	10.12	86.91	86.63	11.8	10.65	16.74	29.11	85.39	99.31
Imazethapyr	4.75	15.84	14.89	11.14	87.7	85.85	7.56	14.63	17.96	12.36	89.51	90.05
Imidacloprid	4.12	13.73	9.63	18.69	82.97	92.52	7.38	26.94	11.85	11.17	90.84	91.09
Iodosulfuron-Methyl	2.87	9.57	6.52	12.32	86.94	85.14	7.49	15.94	14.96	10.12	85.23	86.93
Ioxynil	2.42	8.08	5.36	11.41	93.64	83.96	6.88	21.25	18.74	11.14	90.35	86.71
Iprodione	0.49	1.63	6.89	17.24	85.09	86.97	7.38	19.2	15.65	13.36	90.9	87.3
Isoxaflutole	3.01	10.02	8.47	9.63	101.63	94.06	7.2	27.58	16.32	14.63	89.68	91.93
Kresoxim methyl	5.85	19.5	8.45	8.47	92.88	88.11	3.96	18.87	15.32	15.32	85.02	86.56
Lambda Cyhalothrin	3.26	10.86	7.23	7.85	93.48	85.43	5.94	24.38	14.85	17.45	91.03	95.71
Lenacil	2.4	8.12	9.62	8.24	94.7	90.01	7.2	22.67	17.85	14.11	94.58	91.53
Lufenuron	4.97	16.56	7.21	7.65	92.49	90.03	4.45	33.83	9.32	9.45	92.08	92.44
Malathion	4.14	13.8	8.24	14.85	90.09	93.99	4.24	39.35	6.21	14.65	83.14	93.37
MCPA	4.42	14.75	9.65	17.85	93.67	85.93	5.78	25.2	8.32	17.23	98.84	88.68
Mecoprop	3.51	11.69	8.21	9.32	96.84	87.24	4.8	24.61	8.14	15.21	91.02	96.15
Mefenpyr-diethyl	5.55	18.51	9.32	6.21	90.22	82.44	3.49	24.96	9.12	14.32	85.58	90.69
Mesosulfuron methyl	5.94	19.81	10.65	8.32	83.83	84.01	5.82	22.93	8.14	12.63	77.87	91.93
Mesotrione	3.3	11.01	11.45	8.14	91.1	86.82	4.84	24.58	5.12	14.36	82.08	89.28
Metalaxyl	4.69	15.65	18.63	9.12	90.53	91.51	8.6	24.02	4.12	12.31	82.96	96.41
Metamitron	3.6	12	14.85	14.54	91.53	94.7	6.69	13.19	6.18	11.32	94.33	98.44
Metconazole	4.66	15.54	15.65	13.65	100.83	91.65	4.07	19.79	6.45	9.74	95.57	92.27
Methidathion	5.16	17.19	12.32	14.52	91.07	87.26	4.32	23.99	8.12	4.65	86.59	92.02
Methomyl	4.08	13.59	14.15	18.65	90.46	85.96	4.2	14.85	9.15	6.18	87.72	89.77
Methoxyfenozide	4.13	13.77	14.78	14.65	82.71	89.59	4.75	14.13	9.12	6.45	89.58	96.25
Metolachlor	5.98	19.93	14.56	17.96	90.08	94.47	5.04	19.26	10.12	8.12	87.05	93.4
Metribuzin	1.34	4.47	18.45	11.85	90.99	86.83	3.23	16.01	15.14	9.15	91.54	90.22
Metsulfuron-methyl	3.32	11.07	14.52	14.96	91.16	91.52	2.43	11.64	14.12	9.12	87.12	96.98
Monocrotophos	1.27	4.23	12.54	18.74	87.18	87.68	5.01	19.4	11.1	10.12	86.71	96.66
Monolinuron	1.64	5.46	16.52	15.65	88.97	83.56	6.17	16.12	13.1	15.14	89.99	99.51
Myclobutanil	4.34	14.48	18.25	16.32	99.31	93.36	0.13	28.67	17.23	14.12	86.63	93.42
Nicosulfuron	4.99	16.63	14.65	15.32	90.05	84.52	3.09	22.31	15.21	11.1	85.85	97.25

Pesticides	Cucumber						Grapefruit					
	LOD	LOQ	Precision		Recovery		LOD	LOQ	Precision		Recovery	
			RSD (%)		%				RSD (%)		%	
			20	100	20	100			10	100	10	100
(µg/kg)						(µg/kg)						
Novaluron	3.29	10.95	14.32	14.85	91.09	88.19	3.39	13.56	14.32	13.1	92.52	96.79
Omethoate	1.48	4.93	13.21	17.85	86.93	87.11	2.09	14.41	12.63	10.21	85.14	90.71
Oxadixyl	3.46	11.53	15.36	9.32	86.71	87.44	4.23	14.01	14.36	10.14	83.96	98.12
Oxamyl	1.27	4.23	14.36	6.21	87.3	86.5	2.68	15.82	12.31	9.85	86.97	91.6
Oxyflourfen	4.77	15.91	11.36	8.32	91.93	83.48	4.18	16.79	11.32	15.17	94.06	99.08
Parathion Methyl	1.28	4.28	11.32	8.14	86.56	93.11	5.81	10.75	9.65	11.12	88.11	86.35
Penconazole	5.93	19.77	11.25	9.12	95.71	83.77	3.56	8.1	10.32	12.23	85.43	96.45
Pendimethalin	4.48	14.95	24.32	8.14	91.53	90.2	1.8	16.68	11.85	18.32	90.01	92.42
Permethrin	4.99	16.62	12.36	14.12	92.44	92.26	1.24	20.58	14.65	14.36	90.03	90.57
Phenmedipham	4.38	14.59	11.17	11.12	93.37	92.3	2.33	0.42	17.64	9.15	93.99	90.06
Phenthoate	5.99	19.97	10.12	10.12	88.68	95.18	3.65	10.3	8.65	9.12	85.93	85.16
Phosalone	5.89	19.64	11.14	18.26	96.15	85.34	2.43	11.3	7.65	10.32	87.24	93.31
Phosmet	6.36	21.21	15.36	17.32	90.69	84.19	4.49	6.96	8.32	11.25	82.44	93.71
Pirimicarb	2.38	7.92	18.36	14.32	91.93	93.02	4.58	14.1	9.63	10.87	84.01	86.76
Pirimiphos methyl	4.55	15.17	12.35	18.65	89.28	88.01	2.83	8.92	12.54	9.63	86.82	89.6
Prochloraz	4.03	13.43	14.36	12.74	96.41	90.23	4.01	13.92	14.56	8.35	91.51	102.06
Procymidone	4.22	14.08	17.96	9.12	98.44	87.9	0.65	19.37	17.85	8.65	94.7	95.94
Profenofos	6.06	20.19	11.85	10.11	92.27	91.11	4.95	11.87	19.63	7.65	91.65	100.07
Profoxydim (Clefoxydim)	2.13	7.12	14.96	17.85	92.02	88.23	5.11	5.99	15.36	8.32	87.26	86.06
Prometryn	4.97	16.57	18.74	19.63	89.77	93.85	5.82	4.12	19.36	9.63	85.96	94.31
Propaquizafop	4.81	16.05	15.65	15.36	96.25	87.19	4.9	7.76	17.36	12.54	89.59	92.67
Propargite	5.55	18.48	16.32	19.36	93.4	96.29	2.76	12.16	16.15	14.56	94.47	100.31
Propazine	4.28	14.26	15.32	21.36	90.22	84.29	4.97	8.11	15.17	17.85	86.83	92.13
Propoxycarbazono	3.96	13.21	14.85	10.12	96.98	87.32	2.74	14.97	11.12	39.47	91.52	96.42
Propyconazole	5.7	19.01	17.85	14.25	96.66	84.74	4.61	15.28	12.23	17.96	87.68	87.28
<b>Propyzamide</b>	4.37	14.56	<b>21.36</b>	9.15	99.51	89.42	3.73	9.44	18.32	11.85	83.56	86.31
Prothopos	5.82	19.41	6.21	9.12	93.42	89.52	3.01	13.37	14.36	14.96	93.36	99.09
Pymetrozine	0.17	0.56	8.32	10.12	97.25	85.74	0.52	2.17	9.15	18.74	84.52	92.48
Pyraclostrobin	5.17	17.23	8.14	15.14	96.79	83.32	2.17	16.51	9.12	15.65	88.19	85.9
Pyraflufen-Ethyl	4.21	14.02	9.12	14.12	90.71	92.91	5.36	17.02	6.45	16.32	87.11	82.99
Pyrazophos	6.02	20.06	8.14	11.1	98.12	88.05	4.67	19.41	8.12	15.32	87.44	98.77
Pyridaben	5.82	19.4	10.12	13.1	91.6	90.89	1.09	16.33	9.15	14.85	86.5	98.8
Pyridaphention	5.75	19.16	10.11	10.21	99.08	96.07	3.95	9.2	9.12	17.85	83.48	97.12
Pyridate	5.22	17.39	14.32	10.14	86.35	91.46	3.4	16.57	10.12	9.32	93.11	94.68
Pyriproxyfen	4.57	15.24	10.34	9.85	96.45	86.32	3.46	9.12	15.14	6.21	83.77	85
Quizalofop-P-Ethyl	4.99	16.63	15.32	9.85	92.42	89.45	3.51	15.36	14.12	8.32	90.2	96.72
Rimsulfuron	0.55	1.82	9.98	10.25	90.57	89.54	2.08	12.44	11.1	8.14	92.26	90.67
Sethoxydim	1.25	4.18	8.32	11.14	90.06	91.31	3.21	10.03	13.1	9.12	92.3	90.39
Simozine	2.33	7.75	7.23	14.25	85.16	90.57	2.7	1.72	10.21	8.14	95.18	95.14
Spirodiclofen	4.82	16.08	8.23	18.21	93.31	92.29	3.67	7.22	10.14	10.21	85.34	95.23
Spiroxamine	2.64	8.79	6.12	14.32	93.71	99.81	4.05	17.85	9.85	10.14	84.19	94.61

Pesticides	Cucumber						Grapefruit					
	LOD	LOQ	Precision		Recovery		LOD	LOQ	Precision		Recovery	
			RSD (%)		%				RSD (%)		%	
			20	100	20	100			10	100	10	100
(µg/kg)						(µg/kg)						
Sulfosulfuron	0.66	2.21	14.23	14.32	86.76	101.91	3.53	15.57	8.45	9.85	93.02	101.91
Tau-Fluvalinate	3.29	10.96	15.21	19.32	89.6	94.87	4.68	3.62	10.11	10.12	88.01	100.96
Tebuconazole	5.79	19.29	14.23	10.12	102.06	95.67	3.02	13.16	14.65	14.12	90.23	96.5
Tebufozide	5.25	17.51	9.14	15.14	95.94	104.15	4.6	11.32	17.65	15.17	87.9	104.15
Teflubenzuron	3.91	13.02	8.14	14.12	100.07	100.6	4.83	11.53	12.11	11.52	91.11	100.6
Tepraloxymid	1.69	5.62	7.56	11.1	86.06	103.01	3.06	11.71	17.15	10.54	88.23	103.01
Terbutylazine	3.55	11.82	8.14	13.1	94.31	102.61	5.62	6.94	19.12	11.57	93.85	102.61
Terbutryn	5.82	19.41	8.65	10.21	92.67	95.62	3.88	10.71	13.25	12.65	87.19	93.95
Tetraconazole	4.73	15.77	7.12	10.14	100.31	93.73	3.19	8.99	10.11	13.25	96.29	99.51
Thiabendazole	2.56	8.53	8.45	9.85	92.13	94.54	3.49	12.23	9.85	14.32	84.29	98.75
Thiacloprid	1.57	5.22	8.69	10.12	96.42	99.77	6.3	13.49	8.63	14.58	87.32	99.77
Thiamethoxam	0.70	2.35	6.25	14.12	87.28	101.08	4.33	11.78	7.98	12.69	84.74	101.08
Thifensulfuron-methyl	0.27	0.91	5.12	15.17	86.31	102.61	2.96	15.6	7.63	11.36	89.42	102.61
Thiohanate methyl	2.73	9.09	4.12	11.12	99.09	102.27	3.7	10.05	7.52	10.87	89.52	102.27
Thiometon	4.12	13.72	6.18	12.23	92.48	102.82	1.4	15.35	9.41	9.15	85.74	102.82
Tolyfluanid	3.21	10.68	6.45	18.32	85.9	92.82	4.21	16.11	7.45	8.32	83.32	97.51
Tralkoxydim	3.4	11.33	8.12	14.36	82.99	98.99	4.26	10.2	11.25	9.74	92.91	98.26
Triadimefon	1.75	5.83	9.15	9.15	98.77	99.6	5.78	18.73	10.87	6.32	88.05	99.6
Triadimenol	3.24	10.8	9.12	9.12	98.8	101.72	5.18	12.93	9.63	7.14	90.89	101.72
Triallate	3.59	11.98	10.12	10.12	97.12	101.45	5.14	10.63	8.35	6.32	96.07	101.45
Triasulfuron	0.55	1.84	15.14	15.14	94.68	98.88	3.27	11.64	8.65	7.52	91.46	98.88
Tribenuron-Methyl	0.51	1.7	14.12	14.12	85	101.79	1.79	20.99	7.65	10.21	86.32	101.79
Trichlorfon	0.79	2.64	11.1	11.1	96.72	99.8	3.2	14.42	8.32	10.14	89.45	99.8
Trifloxystrobin	3.6	11.99	13.1	13.1	90.67	102.21	3.04	9.86	9.63	9.85	89.54	102.21
Triflumizole	4.76	15.87	10.21	10.21	90.39	100.93	2.56	12.33	12.54	10.12	91.31	100.93
Triflumuron	3.99	13.31	10.14	10.14	95.14	102.11	1.51	4.67	14.56	14.12	90.57	102.11
Triticonazole	3.19	10.65	9.85	9.85	95.23	101.54	4.63	14.03	17.85	15.17	92.29	101.54
2,4-D Acid	4.39	14.63	7.65	11.23	94.61	102.37	2.07	14.21	14.12	9.12	99.81	102.37

LOD: limit of detection (µg/kg); LOQ: limit of quantification (µg/kg)

Calculated at 20 µg/kg and 100 µg/kg in fortified cucumber matrix

Calculated at 10 µg/kg and 100 µg/kg in fortified grapefruit matrix

The fungicides metalxyl and procymidone were found in 4 cucumber samples at levels varying from 0.011 to 0.039 mg kg<sup>-1</sup> and from 0.014 to 0.033 mg kg<sup>-1</sup>, respectively. The insecticide chlorpyrifos was detectable only in 1 cucumber samples, with a mean concentration of 0.037 mg kg<sup>-1</sup>. Similarly, two cucumber samples were found to contain iprodione, Carbendazim, which is an authorised neonicotinoid insecticide. The concentrations of iprodione, carbendazim in positive samples were from 0.022 to 0.029 mg kg<sup>-1</sup> and from 0.014 to 0.032 mg kg<sup>-1</sup>, respectively.

The results show that grapefruits had slightly higher frequencies (13.8%) of pesticide residues than cucumber samples (11.6%), but the number of residues was less. Of the

233 target analytes monitored, only 9 pesticide residues were found in grapefruits at concentrations varying from 0.002 to 0.042 mg kg<sup>-1</sup>; none above the MRL. Multiple residues in grapefruits were found in 3.5% of the samples. While 15 grapefruit samples contained two residues of pesticides, only 2 samples were contaminated with three pesticide residues.

The fungicide fludioxonil was the most prominent pesticide found in grapefruit samples, with a frequency of 9.1%, and with levels ranging from 0.019 to 0.041 mg kg<sup>-1</sup>. Fludioxonil, which belongs to the class of phenylpyrrole, is used to control a broad spectrum of plant diseases in various vegetables. The acute toxicity of fludioxonil is low. Fludioxonil did not have carcinogenic potential (FAO, 2012).

Imazalil was the second most commonly found pesticide in grapefruits with 4.7% of the samples. The results obtained from the second and third year showed that 7 samples contained imazalil at levels varying from 0.017 to 0.102 mg kg<sup>-1</sup>, and from 0.021 to 0.109 mg kg<sup>-1</sup>, respectively. In the first year, 4 samples had this residue at concentrations ranging from 0.016 to 0.032 mg kg<sup>-1</sup>.

Thiabendazol was detected in 7 samples at levels ranging from 0.014 to 0.081 mg kg<sup>-1</sup>, of which 5 samples contained simultaneously the fungicide quinophos. Thiabendazol, belongs to the class of benzimidazole. It is a systemic fungicide used in citrus fruits such as orange, lemon and grapefruit against *Penicillium digitatum*.

The insecticide carbaryl was present in 4 grapefruit samples, with a mean concentration of 0.028 mg kg<sup>-1</sup>. Carbaryl belongs to the class of carbamate. It is a pesticide and plant growth regulator used in citrus fruits such as orange, lemon, grapefruit.

### ***Uncertainty assessment***

The results of method uncertainty assessment considering six sources are presented in *Table 2*. For cucumber samples the overall contributions to  $U_c$  of the method of the uncertainties associated with  $U_1$  (standard solution preparation),  $U_2$  (calibration curve preparation),  $U_3$  (sample preparation treatment),  $U_4$  (precision), and  $U_5$  (accuracy/bias) ranged from 5.3% to 10.6% and  $U_6$  (linear least square fitting) ranged from 85.6% to 93.1%. In the same way, for grapefruit, the overall contributions of the uncertainties associated with  $U_1$ ,  $U_2$ ,  $U_3$ ,  $U_4$ , and  $U_5$  varied from 5.1% to 11.8% and  $U_6$  ranged from 87.8% to 95.1%. In the light of these results, the uncertainty associated with  $u_6$  presents the highest contribution for all the pesticides under study in cucumber and grapefruit.

As can be seen in *Table 2*,  $U_c$  values at 95% confidence level for cucumber ranged from 12.3% to 18.7% and for grapefruit from 15.6% to 23.6%.  $U_c$  values of all pesticides detected in cucumber samples generally offer lower values than grapefruit. For all target analytes, the  $U_c$  was within the criteria specified in the SANTE Guideline (less than 50%). These results are similar to those reported for methods using GC-MS, LCMS-MS and the QuEChERS procedure. For example, Walorczyk (2014) reported  $U_c$  values ranging from 7.0% to 53.0% for samples of black currant, where the majority of pesticides had a uncertainty of <30.0%. In addition, da Silva Sousa et al. (2018) reported  $U_c$  values from 7.9% to 36.1% for melon samples. At first glance, the results of this method are high, but the uncertainty acceptance criteria according to the European SANTE / 11945/2015 guideline European Commission (2015) is the time when the value is 50.0%. Although it is recommended to use the internal standard to minimize uncertainties, the proposed method indicated that expanded uncertainties are not necessary.

**Table 2.** Method uncertainties estimated for different sources and, combined ( $U_c$ ) and expanded ( $U_e$ ) uncertainty

Pesticide	Cucumber										Grapefruit									
	$U_1$	$U_2$	$U_i^a$ (mg/kg (%))		$U_5$	$U_6$	$U_c$	$U_e^b$	$U_e(\%)$		$U_1$	$U_2$	$U_i^a$ (mg/kg (%))		$U_5$	$U_6$	$U_c$	$U_e^b$	$U_e(\%)$	
			$U_3$	$U_4$									$U_3$	$U_4$						
Trifluralin	0.044 (2.3)	0.036 (1.0)	0.031 (0.9)	0.023 (0.7)	0.039 (1.8)	0.301 (92.1)	0.313	0.618	15.6		0.044 (2.3)	0.036 (1.0)	0.031 (0.9)	0.050 (1.7)	0.064 (2.8)	0.350 (88.4)	0.384	0.765	15.7	
Chlorothalonil	0.044 (2.1)	0.036 (0.9)	0.031 (0.8)	0.072 (4.3)	0.032 (1.5)	0.317 (89.5)	0.311	0.662	15.8		0.044 (2.1)	0.036 (0.9)	0.031 (0.8)	0.034 (1.2)	0.044 (1.7)	0.318 (87.8)	0.382	0.665	16.4	
Formothion	0.044 (2.0)	0.036 (1.2)	0.031 (1.5)	0.008 (4.2)	0.054 (2.6)	0.245 (90.4)	0.320	0.665	16.2		0.044 (2.0)	0.036 (1.2)	0.031 (1.5)	0.076 (2.2)	0.068 (1.6)	0.333 (92.1)	0.381	0.654	18.3	
Vinclozolin	0.044 (3.2)	0.036 (1.1)	0.031 (1.6)	0.007 (4.1)	0.048 (2.2)	0.298 (88.7)	0.254	0.514	16.3		0.044 (3.2)	0.036 (1.1)	0.031 (1.6)	0.071 (4.4)	0.060 (3.5)	0.368 (91.2)	0.504	0.774	19.5	
Fenitrothion	0.044 (3.5)	0.036 (1.5)	0.031 (1.0)	0.009 (3.9)	0.038 (1.8)	0.332 (85.6)	0.256	0.512	15.6		0.044 (3.5)	0.036 (1.5)	0.031 (1.0)	0.097 (6.3)	0.071 (3.4)	0.495 (95.1)	0.501	0.771	15.6	
Dicofol	0.044 (1.9)	0.036 (1.6)	0.031 (1.1)	0.070 (3.7)	0.039 (1.8)	0.312 (92.4)	0.245	0.628	12.6		0.044 (1.9)	0.036 (1.6)	0.031 (1.1)	0.096 (5.4)	0.062 (2.6)	0.452 (94.1)	0.359	0.659	16.2	
Captan	0.044 (2.0)	0.036 (1.7)	0.031 (1.4)	0.066 (0.2)	0.041 (2.1)	0.298 (89.7)	0.287	0.624	12.8		0.044 (2.0)	0.036 (1.7)	0.031 (1.4)	0.071 (2.1)	0.045 (1.6)	0.413 (92.1)	0.385	0.662	19.6	
Folpet	0.044 (3.4)	0.036 (1.2)	0.031 (1.3)	0.052 (0.1)	0.068 (3.5)	0.291 (93.1)	0.298	0.701	12.9		0.044 (3.4)	0.036 (1.2)	0.031 (1.3)	0.033 (1.5)	0.061 (3.2)	0.402 (90.8)	0.504	0.652	20.3	
Qinomethionate	0.044 (3.3)	0.036 (1.4)	0.031 (0.9)	0.054 (0.6)	0.071 (3.9)	0.335 (95.4)	0.301	0.704	12.7		0.044 (3.3)	0.036 (1.4)	0.031 (0.9)	0.051 (1.9)	0.070 (3.3)	0.350 (88.4)	0.501	1.014	21.6	
Dinobuton	0.044 (3.2)	0.036 (1.5)	0.031 (1.1)	0.051 (0.7)	0.065 (4.1)	0.301 (92.1)	0.305	0.517	12.8		0.044 (3.2)	0.036 (1.5)	0.031 (1.1)	0.076 (2.2)	0.064 (2.8)	0.318 (87.8)	0.328	0.665	23.4	
Endosulfan A	0.044 (3.1)	0.036 (1.2)	0.031 (1.4)	0.041 (0.5)	0.070 (3.6)	0.317 (89.5)	0.321	0.514	13.1		0.044 (3.1)	0.036 (1.2)	0.031 (1.4)	0.071 (4.4)	0.044 (1.7)	0.333 (92.1)	0.329	1.011	21.8	
Endosulfan B	0.044 (2.1)	0.036 (1.3)	0.031 (1.3)	0.040 (0.6)	0.068 (4.2)	0.245 (90.4)	0.315	0.548	13.6		0.044 (2.1)	0.036 (1.3)	0.031 (1.3)	0.097 (6.3)	0.068 (1.6)	0.368 (91.2)	0.385	0.774	22.8	
Bromopropylate	0.044 (3.5)	0.036 (1.0)	0.031 (0.9)	0.043 (0.2)	0.039 (1.8)	0.298 (88.7)	0.311	0.621	13.8		0.044 (3.5)	0.036 (1.0)	0.031 (0.9)	0.096 (5.4)	0.060 (3.5)	0.495 (95.1)	0.384	0.776	21.8	
Tetradifon	0.044 (3.3)	0.036 (0.9)	0.031 (0.7)	0.054 (0.4)	0.048 (2.2)	0.332 (85.6)	0.314	0.514	13.9		0.044 (3.3)	0.036 (0.9)	0.031 (0.7)	0.071 (2.1)	0.071 (3.4)	0.452 (94.1)	0.391	0.778	23.6	
Alfa-Cypermethrin a	0.044 (2.3)	0.036 (1.2)	0.031 (0.8)	0.071 (0.3)	0.038 (1.8)	0.312 (92.4)	0.317	0.665	14.6		0.044 (2.3)	0.036 (1.2)	0.031 (0.8)	0.033 (1.5)	0.062 (2.6)	0.413 (92.1)	0.398	0.662	20.8	
Alfa-Cypermethrin b	0.044 (2.1)	0.036 (1.1)	0.031 (0.9)	0.041 (0.5)	0.039 (1.8)	0.298 (89.7)	0.298	0.541	15.8		0.044 (2.1)	0.036 (1.1)	0.031 (0.9)	0.051 (1.9)	0.045 (1.6)	0.402 (90.8)	0.387	1.010	19.6	
alpha.-BHC	0.044 (2.0)	0.036 (1.5)	0.031 (1.0)	0.040 (0.6)	0.041 (2.1)	0.291 (93.1)	0.289	0.526	14.9		0.044 (2.0)	0.036 (1.5)	0.031 (1.0)	0.076 (2.2)	0.061 (3.2)	0.350 (88.4)	0.384	0.669	18.9	
delta.-BHC	0.044 (3.2)	0.036 (1.6)	0.031 (1.1)	0.043 (0.2)	0.068 (3.5)	0.331 (91.7)	0.287	0.668	15.6		0.044 (3.2)	0.036 (1.6)	0.031 (1.1)	0.071 (4.4)	0.070 (3.3)	0.318 (87.8)	0.385	0.771	17.6	
Heptachlor	0.044 (3.5)	0.036 (1.7)	0.031 (1.4)	0.054 (0.4)	0.032 (1.5)	0.301 (92.1)	0.269	0.662	16.9		0.044 (3.5)	0.036 (1.7)	0.031 (1.4)	0.097 (6.3)	0.064 (2.8)	0.333 (92.1)	0.381	0.778	15.9	
2,4-DDE	0.044 (1.9)	0.036 (1.4)	0.031 (1.3)	0.071 (0.3)	0.039 (1.8)	0.317 (89.5)	0.278	0.669	16.8		0.044 (1.9)	0.036 (1.4)	0.031 (1.3)	0.096 (5.4)	0.044 (1.7)	0.368 (91.2)	0.329	0.668	16.3	

Pesticide	Cucumber									Grapefruit								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)
			U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>							U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>				
(mg/kg)									(mg/kg)									
Dieldrin	0.044 (2.0)	0.036 (1.5)	0.031 (0.9)	0.072 (4.3)	0.045 (2.7)	0.245 (90.4)	0.259	0.554	14.3	0.044 (2.0)	0.036 (1.5)	0.031 (0.9)	0.071 (2.1)	0.068 (1.6)	0.495 (95.1)	0.501	0.669	15.9
4,4-DDE	0.044 (3.4)	0.036 (1.2)	0.031 (0.7)	0.008 (4.2)	0.065 (2.6)	0.298 (88.7)	0.314	0.552	15.9	0.044 (3.4)	0.036 (1.2)	0.031 (0.7)	0.033 (1.5)	0.060 (3.5)	0.452 (94.1)	0.384	0.765	18.6
2,4-DDD	0.044 (3.3)	0.036 (1.3)	0.031 (0.8)	0.007 (4.1)	0.048 (2.2)	0.332 (85.6)	0.315	0.661	17.6	0.044 (3.3)	0.036 (1.3)	0.031 (0.8)	0.051 (1.9)	0.071 (3.4)	0.413 (92.1)	0.382	0.665	18.7
Endrin	0.044 (3.2)	0.036 (1.0)	0.031 (0.9)	0.009 (3.9)	0.038 (1.8)	0.312 (92.4)	0.347	0.618	18.6	0.044 (3.2)	0.036 (1.0)	0.031 (0.9)	0.097 (6.3)	0.062 (2.6)	0.402 (90.8)	0.381	0.654	16.9
Chlorfenapyf	0.044 (3.1)	0.036 (0.8)	0.031 (1.0)	0.070 (3.7)	0.039 (1.8)	0.291 (93.1)	0.325	0.662	17.8	0.044 (3.1)	0.036 (0.8)	0.031 (1.0)	0.096 (5.4)	0.045 (1.6)	0.350 (88.4)	0.504	0.774	18.3
4,4-DDD	0.044 (2.1)	0.036 (1.0)	0.031 (1.1)	0.066 (0.2)	0.041 (2.1)	0.330 (94.5)	0.287	0.665	17.6	0.044 (2.1)	0.036 (1.0)	0.031 (1.1)	0.071 (2.1)	0.061 (3.2)	0.318 (87.8)	0.501	0.771	16.9
Ethion	0.044 (3.4)	0.036 (0.9)	0.031 (1.4)	0.052 (0.1)	0.068 (3.5)	0.296 (95.2)	0.298	0.514	15.9	0.044 (3.4)	0.036 (0.9)	0.031 (1.4)	0.033 (1.5)	0.070 (3.3)	0.333 (92.1)	0.359	0.659	18.1
DDT	0.044 (1.9)	0.036 (1.2)	0.031 (1.3)	0.054 (0.6)	0.052 (3.4)	0.332 (85.6)	0.301	0.512	18.1	0.044 (1.9)	0.036 (1.2)	0.031 (1.3)	0.051 (1.9)	0.064 (2.8)	0.368 (91.2)	0.385	0.662	18.6
Abamectin	0.044 (2.1)	0.036 (1.1)	0.031 (0.9)	0.051 (0.7)	0.037 (1.7)	0.301 (92.1)	0.305	0.628	17.6	0.044 (2.1)	0.036 (1.1)	0.031 (0.9)	0.076 (2.2)	0.044 (1.7)	0.495 (95.1)	0.504	0.652	17.8
Acetamiprid	0.044 (2.3)	0.036 (1.5)	0.031 (1.1)	0.041 (0.5)	0.045 (1.8)	0.317 (89.5)	0.321	0.624	15.9	0.044 (2.3)	0.036 (1.5)	0.031 (1.1)	0.071 (4.4)	0.068 (1.6)	0.452 (94.1)	0.501	1.014	17.3
Acetochlor	0.044 (2.0)	0.036 (1.6)	0.031 (1.4)	0.040 (0.6)	0.065 (2.6)	0.245 (90.4)	0.315	0.701	14.3	0.044 (2.0)	0.036 (1.6)	0.031 (1.4)	0.097 (6.3)	0.060 (3.5)	0.413 (92.1)	0.328	0.665	19.6
Acrinathrin	0.044 (3.4)	0.036 (1.7)	0.031 (1.3)	0.043 (0.2)	0.048 (2.2)	0.298 (88.7)	0.311	0.704	13.6	0.044 (3.4)	0.036 (1.7)	0.031 (1.3)	0.096 (5.4)	0.071 (3.4)	0.402 (90.8)	0.329	1.011	20.5
Alachlor	0.044 (3.3)	0.036 (1.4)	0.031 (0.9)	0.054 (0.4)	0.062 (2.1)	0.332 (85.6)	0.314	0.517	13.5	0.044 (3.3)	0.036 (1.4)	0.031 (0.9)	0.071 (2.1)	0.062 (2.6)	0.350 (88.4)	0.385	0.774	21.5
Amitraz	0.044 (3.2)	0.036 (1.5)	0.031 (0.7)	0.071 (0.3)	0.039 (1.8)	0.312 (92.4)	0.317	0.514	18.1	0.044 (3.2)	0.036 (1.5)	0.031 (0.7)	0.033 (1.5)	0.045 (1.6)	0.318 (87.8)	0.384	0.776	22.5
Atrazine	0.044 (3.1)	0.036 (1.2)	0.031 (0.8)	0.041 (0.5)	0.065 (2.6)	0.298 (89.7)	0.298	0.548	14.5	0.044 (3.1)	0.036 (1.2)	0.031 (0.8)	0.051 (1.9)	0.061 (3.2)	0.333 (92.1)	0.507	0.669	21.7
Azinphos methyl	0.044 (2.1)	0.036 (1.3)	0.031 (0.9)	0.040 (0.6)	0.048 (2.2)	0.291 (93.1)	0.298	0.664	16.5	0.044 (2.1)	0.036 (1.3)	0.031 (0.9)	0.076 (2.2)	0.070 (3.3)	0.368 (91.2)	0.504	0.771	22.9
Azoxystrobin	0.044 (2.1)	0.036 (1.0)	0.031 (1.0)	0.043 (0.2)	0.048 (2.2)	0.332 (85.6)	0.337	0.663	15.7	0.044 (2.1)	0.036 (1.0)	0.031 (1.0)	0.071 (4.4)	0.064 (2.8)	0.495 (95.1)	0.394	0.778	21.7
Bensulfuron-Methyl	0.044 (2.0)	0.036 (0.8)	0.031 (1.1)	0.054 (0.4)	0.038 (1.8)	0.301 (92.1)	0.315	0.559	14.9	0.044 (2.0)	0.036 (0.8)	0.031 (1.1)	0.097 (6.3)	0.044 (1.7)	0.452 (94.1)	0.392	0.668	20.6
Bentazone	0.044 (3.2)	0.036 (0.9)	0.031 (1.4)	0.071 (0.3)	0.039 (1.8)	0.317 (89.5)	0.347	0.557	17.6	0.044 (3.2)	0.036 (0.9)	0.031 (1.4)	0.096 (5.4)	0.068 (1.6)	0.413 (92.1)	0.389	0.669	15.7
Beta-Cyfluthrin	0.044 (3.5)	0.036 (1.0)	0.031 (1.3)	0.041 (0.5)	0.041 (2.1)	0.245 (90.4)	0.325	0.621	18.1	0.044 (3.5)	0.036 (1.0)	0.031 (1.3)	0.071 (2.1)	0.060 (3.5)	0.402 (90.8)	0.374	1.015	16.4
Bifenthrin	0.044 (1.9)	0.036 (0.9)	0.031 (0.9)	0.040 (0.6)	0.068 (3.5)	0.298 (88.7)	0.289	0.514	16.9	0.044 (1.9)	0.036 (0.9)	0.031 (0.9)	0.033 (1.5)	0.071 (3.4)	0.350 (88.4)	0.365	1.016	18.3

Pesticide	Cucumber									Grapefruit								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)
			U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>							U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>				
(mg/kg)									(mg/kg)									
Boscalid	0.044 (2.0)	0.036 (1.2)	0.031 (0.7)	0.043 (0.2)	0.065 (2.6)	0.332 (85.6)	0.287	0.665	18.2	0.044 (2.0)	0.036 (1.2)	0.031 (0.7)	0.051 (1.9)	0.062 (2.6)	0.318 (87.8)	0.361	0.887	19.5
Bromoxynil	0.044 (3.4)	0.036 (1.1)	0.031 (0.8)	0.054 (0.4)	0.048 (2.2)	0.312 (92.4)	0.269	0.541	14.3	0.044 (3.4)	0.036 (1.1)	0.031 (0.8)	0.076 (2.2)	0.045 (1.6)	0.333 (92.1)	0.368	0.778	15.6
Bromuconazole	0.044 (3.3)	0.036 (1.5)	0.031 (0.9)	0.071 (0.3)	0.062 (2.1)	0.298 (89.7)	0.278	0.526	15.1	0.044 (3.3)	0.036 (1.5)	0.031 (0.9)	0.071 (4.4)	0.061 (3.2)	0.368 (91.2)	0.501	0.779	16.2
Bupirimate	0.044 (3.2)	0.036 (1.6)	0.031 (1.0)	0.008 (4.2)	0.045 (1.8)	0.291 (93.1)	0.259	0.668	18.3	0.044 (3.2)	0.036 (1.6)	0.031 (1.0)	0.097 (6.3)	0.070 (3.3)	0.495 (95.1)	0.500	0.765	19.6
Buprofezine	0.044 (3.1)	0.036 (1.7)	0.031 (1.4)	0.007 (4.1)	0.048 (2.2)	0.332 (85.6)	0.335	0.662	14.6	0.044 (3.1)	0.036 (1.7)	0.031 (1.4)	0.096 (5.4)	0.064 (2.8)	0.452 (94.1)	0.384	0.669	20.3
Carbaryl	0.044 (2.1)	0.036 (1.4)	0.031 (1.3)	0.009 (3.9)	0.038 (1.8)	0.301 (92.1)	0.287	0.669	17.6	0.044 (2.1)	0.036 (1.4)	0.031 (1.3)	0.071 (2.1)	0.044 (1.7)	0.413 (92.1)	0.382	0.765	21.6
Carbendazim	0.044 (2.3)	0.036 (1.5)	0.031 (0.9)	0.070 (3.7)	0.039 (1.8)	0.317 (89.5)	0.298	0.665	15.9	0.044 (2.3)	0.036 (1.5)	0.031 (0.9)	0.033 (1.5)	0.068 (1.6)	0.402 (90.8)	0.381	0.665	23.4
Carbofuran	0.044 (2.1)	0.036 (1.2)	0.031 (1.1)	0.066 (0.2)	0.041 (2.1)	0.245 (90.4)	0.301	0.514	17.6	0.044 (2.1)	0.036 (1.2)	0.031 (1.1)	0.051 (1.9)	0.060 (3.5)	0.350 (88.4)	0.504	0.654	21.8
Carbosulfan	0.044 (1.9)	0.036 (1.3)	0.031 (1.4)	0.052 (0.1)	0.068 (3.5)	0.298 (88.7)	0.305	0.512	18.6	0.044 (1.9)	0.036 (1.3)	0.031 (1.4)	0.034 (1.2)	0.071 (3.4)	0.318 (87.8)	0.501	0.774	22.8
Carboxin	0.044 (2.3)	0.036 (1.0)	0.031 (1.3)	0.054 (0.6)	0.052 (3.4)	0.291 (93.1)	0.321	0.628	17.8	0.044 (2.3)	0.036 (1.0)	0.031 (1.3)	0.076 (2.2)	0.062 (2.6)	0.333 (92.1)	0.359	0.771	21.8
Carfentrazone-Ethyl	0.044 (2.3)	0.036 (0.9)	0.031 (0.9)	0.051 (0.7)	0.037 (1.7)	0.338 (89.6)	0.315	0.624	17.6	0.044 (2.3)	0.036 (0.9)	0.031 (0.9)	0.071 (4.4)	0.045 (1.6)	0.368 (91.2)	0.385	0.659	23.6
Chlofentazine	0.044 (2.1)	0.036 (1.0)	0.031 (0.7)	0.041 (0.5)	0.045 (1.8)	0.332 (85.6)	0.311	0.701	15.9	0.044 (2.1)	0.036 (1.0)	0.031 (0.7)	0.097 (6.3)	0.061 (3.2)	0.495 (95.1)	0.504	0.662	20.8
Chlorfluozuron	0.044 (2.0)	0.036 (0.9)	0.031 (0.8)	0.040 (0.6)	0.062 (2.1)	0.312 (92.4)	0.314	0.704	18.1	0.044 (2.0)	0.036 (0.9)	0.031 (0.8)	0.096 (5.4)	0.070 (3.3)	0.452 (94.1)	0.501	0.652	19.6
Chloridazon	0.044 (3.2)	0.036 (1.2)	0.031 (0.9)	0.043 (0.2)	0.039 (1.8)	0.298 (89.7)	0.317	0.517	17.6	0.044 (3.2)	0.036 (1.2)	0.031 (0.9)	0.071 (2.1)	0.064 (2.8)	0.413 (92.1)	0.328	1.014	18.9
Chlormequat chloride	0.044 (3.5)	0.036 (1.1)	0.031 (1.0)	0.054 (0.4)	0.065 (2.6)	0.301 (92.1)	0.298	0.514	15.9	0.044 (3.5)	0.036 (1.1)	0.031 (1.0)	0.033 (1.5)	0.044 (1.7)	0.402 (90.8)	0.329	0.665	17.6
Chlorpham	0.044 (1.9)	0.036 (1.5)	0.031 (1.6)	0.071 (0.3)	0.048 (2.2)	0.317 (89.5)	0.331	0.548	14.9	0.044 (1.9)	0.036 (1.5)	0.031 (1.6)	0.051 (1.9)	0.068 (1.6)	0.350 (88.4)	0.385	1.011	15.9
Chlorpyrifos ethyl	0.044 (2.0)	0.036 (1.6)	0.031 (1.0)	0.041 (0.5)	0.045 (1.8)	0.245 (90.4)	0.289	0.514	17.5	0.044 (2.0)	0.036 (1.6)	0.031 (1.0)	0.076 (2.2)	0.060 (3.5)	0.318 (87.8)	0.384	0.774	19.8
Chlorpyrifos Methyl	0.044 (3.4)	0.036 (1.7)	0.031 (1.1)	0.040 (0.6)	0.048 (2.2)	0.298 (88.7)	0.287	0.665	15.3	0.044 (3.4)	0.036 (1.7)	0.031 (1.1)	0.071 (4.4)	0.071 (3.4)	0.333 (92.1)	0.394	0.776	18.7
Chlorsulfuron	0.044 (3.3)	0.036 (1.4)	0.031 (1.4)	0.043 (0.2)	0.038 (1.8)	0.291 (93.1)	0.269	0.541	16.1	0.044 (3.3)	0.036 (1.4)	0.031 (1.4)	0.097 (6.3)	0.062 (2.6)	0.368 (91.2)	0.392	0.669	19.3
Clethodim	0.044 (3.2)	0.036 (1.5)	0.031 (1.3)	0.054 (0.4)	0.039 (1.8)	0.298 (89.7)	0.278	0.526	14.7	0.044 (3.2)	0.036 (1.5)	0.031 (1.3)	0.096 (5.4)	0.045 (1.6)	0.495 (95.1)	0.389	0.771	17.6

Pesticide	Cucumber									Grapefruit								
	U <sub>i</sub> <sup>a</sup> (mg/kg (%))									U <sub>i</sub> <sup>a</sup> (mg/kg (%))								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)
(mg/kg)									(mg/kg)									
Clodinafop-Propargyl	0.044 (3.1)	0.036 (1.2)	0.031 (0.9)	0.071 (0.3)	0.041 (2.1)	0.332 (85.6)	0.259	0.668	15.2	0.044 (3.1)	0.036 (1.2)	0.031 (0.9)	0.071 (2.1)	0.061 (3.2)	0.452 (94.1)	0.374	0.778	20.1
Clothianidin	0.044 (2.1)	0.036 (1.3)	0.031 (1.1)	0.008 (4.2)	0.068 (3.5)	0.312 (92.4)	0.332	0.662	12.5	0.044 (2.1)	0.036 (1.3)	0.031 (1.1)	0.033 (1.5)	0.070 (3.3)	0.413 (92.1)	0.365	0.668	21.5
Cyclanilide	0.044 (2.3)	0.036 (1.0)	0.031 (1.4)	0.007 (4.1)	0.065 (2.6)	0.298 (89.7)	0.287	0.669	13.6	0.044 (2.3)	0.036 (1.0)	0.031 (1.4)	0.051 (1.9)	0.064 (2.8)	0.402 (90.8)	0.361	0.669	23.4
Cycloate	0.044 (2.1)	0.036 (0.8)	0.031 (1.3)	0.009 (3.9)	0.048 (2.2)	0.301 (92.1)	0.298	0.665	18.1	0.044 (2.1)	0.036 (0.8)	0.031 (1.3)	0.034 (1.2)	0.044 (1.7)	0.350 (88.4)	0.368	0.664	20.9
Cyhalofop-butyl	0.044 (1.9)	0.036 (0.9)	0.031 (0.9)	0.070 (3.7)	0.039 (1.8)	0.317 (89.5)	0.301	0.514	14.8	0.044 (1.9)	0.036 (0.9)	0.031 (0.9)	0.076 (2.2)	0.068 (1.6)	0.318 (87.8)	0.498	0.774	21.7
Cymoxanil	0.044 (3.2)	0.036 (1.0)	0.031 (0.7)	0.066 (0.2)	0.062 (2.1)	0.245 (90.4)	0.305	0.512	17.5	0.044 (3.2)	0.036 (1.0)	0.031 (0.7)	0.071 (4.4)	0.060 (3.5)	0.333 (92.1)	0.465	0.778	19.8
Cypermethrin	0.044 (3.1)	0.036 (0.9)	0.031 (0.8)	0.052 (0.1)	0.048 (2.2)	0.298 (88.7)	0.321	0.628	17.6	0.044 (3.1)	0.036 (0.9)	0.031 (0.8)	0.097 (6.3)	0.071 (3.4)	0.368 (91.2)	0.395	0.779	19.5
Cyproconazole	0.044 (2.8)	0.036 (1.2)	0.031 (0.9)	0.054 (0.6)	0.038 (1.8)	0.337 (92.6)	0.315	0.624	12.5	0.044 (2.8)	0.036 (1.2)	0.031 (0.9)	0.096 (5.4)	0.062 (2.6)	0.495 (95.1)	0.401	1.010	19.1
Cyprodinil	0.044 (2.0)	0.036 (1.1)	0.031 (1.0)	0.051 (0.7)	0.039 (1.8)	0.298 (89.7)	0.311	0.701	14.8	0.044 (2.0)	0.036 (1.1)	0.031 (1.0)	0.071 (2.1)	0.045 (1.6)	0.452 (94.1)	0.425	1.011	18.7
Dazomet	0.044 (3.4)	0.036 (1.5)	0.031 (1.1)	0.041 (0.5)	0.041 (2.1)	0.332 (85.6)	0.314	0.704	17.9	0.044 (3.4)	0.036 (1.5)	0.031 (1.1)	0.033 (1.5)	0.061 (3.2)	0.413 (92.1)	0.384	0.998	19.3
Deltamethrin	0.044 (3.3)	0.036 (1.6)	0.031 (1.4)	0.040 (0.6)	0.068 (3.5)	0.312 (92.4)	0.317	0.517	18.6	0.044 (3.3)	0.036 (1.6)	0.031 (1.4)	0.051 (1.9)	0.070 (3.3)	0.402 (90.8)	0.382	0.884	16.4
Diafenthiuron	0.044 (3.2)	0.036 (1.7)	0.031 (1.3)	0.043 (0.2)	0.039 (1.8)	0.298 (89.7)	0.298	0.514	16.5	0.044 (3.2)	0.036 (1.7)	0.031 (1.3)	0.076 (2.2)	0.064 (2.8)	0.350 (88.4)	0.381	0.765	19.1
Diazinon	0.044 (3.1)	0.036 (1.4)	0.031 (0.9)	0.054 (0.4)	0.065 (2.6)	0.301 (92.1)	0.289	0.548	15.4	0.044 (3.1)	0.036 (1.4)	0.031 (0.9)	0.071 (4.4)	0.044 (1.7)	0.318 (87.8)	0.504	0.665	18.4
Dicamba	0.044 (2.1)	0.036 (1.5)	0.031 (0.7)	0.071 (0.3)	0.048 (2.2)	0.317 (89.5)	0.287	0.612	15.8	0.044 (2.1)	0.036 (1.5)	0.031 (0.7)	0.097 (6.3)	0.068 (1.6)	0.333 (92.1)	0.501	0.654	19.6
Dichlofluanid	0.044 (2.1)	0.036 (1.2)	0.031 (0.8)	0.041 (0.5)	0.048 (2.2)	0.245 (90.4)	0.269	0.514	15.9	0.044 (2.1)	0.036 (1.2)	0.031 (0.8)	0.096 (5.4)	0.060 (3.5)	0.368 (91.2)	0.359	0.774	18.9
Dichlorvos	0.044 (2.0)	0.036 (1.3)	0.031 (0.9)	0.040 (0.6)	0.038 (1.8)	0.298 (88.7)	0.278	0.665	17.6	0.044 (2.0)	0.036 (1.3)	0.031 (0.9)	0.071 (2.1)	0.071 (3.4)	0.495 (95.1)	0.385	0.771	15.8
Diclofop Methyl	0.044 (3.2)	0.036 (1.0)	0.031 (1.0)	0.043 (0.2)	0.039 (1.8)	0.332 (85.6)	0.259	0.541	18.6	0.044 (3.2)	0.036 (1.0)	0.031 (1.0)	0.033 (1.5)	0.062 (2.6)	0.452 (94.1)	0.504	0.659	19.6
Diethofencarb	0.044 (3.5)	0.036 (1.6)	0.031 (1.4)	0.054 (0.4)	0.041 (2.1)	0.312 (92.4)	0.315	0.526	17.8	0.044 (3.5)	0.036 (1.6)	0.031 (1.4)	0.051 (1.9)	0.045 (1.6)	0.413 (92.1)	0.501	0.662	20.1
Difenoconazole	0.044 (1.9)	0.036 (1.5)	0.031 (1.3)	0.071 (0.3)	0.068 (3.5)	0.298 (89.7)	0.347	0.668	17.6	0.044 (1.9)	0.036 (1.5)	0.031 (1.3)	0.034 (1.2)	0.061 (3.2)	0.402 (90.8)	0.328	0.652	15.7
Difflubenzuron	0.044 (3.2)	0.036 (1.0)	0.031 (0.9)	0.007 (4.1)	0.065 (2.6)	0.291 (93.1)	0.325	0.662	15.9	0.044 (3.2)	0.036 (1.0)	0.031 (0.9)	0.076 (2.2)	0.070 (3.3)	0.350 (88.4)	0.329	1.014	16.4
Dimethenamid	0.044 (3.1)	0.036 (0.9)	0.031 (1.1)	0.009 (3.9)	0.048 (2.2)	0.337 (92.6)	0.333	0.669	18.1	0.044 (3.1)	0.036 (0.9)	0.031 (1.1)	0.071 (4.4)	0.064 (2.8)	0.318 (87.8)	0.385	0.665	18.3

Pesticide	Cucumber									Grapefruit								
	U <sub>i</sub> <sup>a</sup> (mg/kg (%))									U <sub>i</sub> <sup>a</sup> (mg/kg (%))								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)
(mg/kg)									(mg/kg)									
Dimethoate	0.044 (2.8)	0.036 (1.2)	0.031 (1.4)	0.070 (3.7)	0.039 (1.8)	0.298 (89.7)	0.287	0.618	17.6	0.044 (2.8)	0.036 (1.2)	0.031 (1.4)	0.097 (6.3)	0.044 (1.7)	0.333 (92.1)	0.384	1.011	19.5
Dimethomorph	0.044 (3.4)	0.036 (1.1)	0.031 (1.3)	0.066 (0.2)	0.062 (2.1)	0.301 (92.1)	0.298	0.662	15.9	0.044 (3.4)	0.036 (1.1)	0.031 (1.3)	0.096 (5.4)	0.068 (1.6)	0.368 (91.2)	0.394	0.774	15.6
Diniconazole	0.044 (3.3)	0.036 (1.5)	0.031 (0.9)	0.052 (0.1)	0.045 (1.8)	0.317 (89.5)	0.301	0.665	15.6	0.044 (3.3)	0.036 (1.5)	0.031 (0.9)	0.071 (2.1)	0.060 (3.5)	0.495 (95.1)	0.392	0.776	16.2
Dinocap	0.044 (3.2)	0.036 (1.6)	0.031 (0.7)	0.054 (0.6)	0.048 (2.2)	0.245 (90.4)	0.305	0.514	15.8	0.044 (3.2)	0.036 (1.6)	0.031 (0.7)	0.033 (1.5)	0.071 (3.4)	0.452 (94.1)	0.389	0.669	19.6
Dioxathion	0.044 (3.1)	0.036 (1.7)	0.031 (0.8)	0.051 (0.7)	0.038 (1.8)	0.298 (88.7)	0.321	0.512	17.4	0.044 (3.1)	0.036 (1.7)	0.031 (0.8)	0.051 (1.9)	0.062 (2.6)	0.413 (92.1)	0.374	0.771	20.3
Diphenamid	0.044 (2.1)	0.036 (1.4)	0.031 (0.9)	0.041 (0.5)	0.039 (1.8)	0.332 (85.6)	0.315	0.628	16.5	0.044 (2.1)	0.036 (1.4)	0.031 (0.9)	0.076 (2.2)	0.045 (1.6)	0.402 (90.8)	0.365	0.778	21.6
Dithianon	0.044 (2.3)	0.036 (1.5)	0.031 (1.0)	0.040 (0.6)	0.041 (2.1)	0.312 (92.4)	0.311	0.624	13.4	0.044 (2.3)	0.036 (1.5)	0.031 (1.0)	0.071 (4.4)	0.061 (3.2)	0.350 (88.4)	0.361	0.668	23.4
Diuron	0.044 (2.1)	0.036 (1.2)	0.031 (1.5)	0.043 (0.2)	0.068 (3.5)	0.298 (89.7)	0.314	0.701	15.2	0.044 (2.1)	0.036 (1.2)	0.031 (1.5)	0.097 (6.3)	0.070 (3.3)	0.318 (87.8)	0.368	0.669	21.8
Epoxyconazole	0.044 (2.0)	0.036 (1.3)	0.031 (1.6)	0.054 (0.4)	0.052 (3.4)	0.337 (92.6)	0.317	0.704	14.2	0.044 (2.0)	0.036 (1.3)	0.031 (1.6)	0.096 (5.4)	0.064 (2.8)	0.333 (92.1)	0.432	0.774	22.8
EPTC	0.044 (3.2)	0.036 (1.0)	0.031 (1.0)	0.071 (0.3)	0.037 (1.7)	0.332 (87.4)	0.298	0.517	18.1	0.044 (3.2)	0.036 (1.0)	0.031 (1.0)	0.071 (2.1)	0.044 (1.7)	0.368 (91.2)	0.425	0.885	21.8
Esfenvalerate	0.044 (3.5)	0.036 (1.4)	0.031 (1.1)	0.041 (0.5)	0.045 (1.8)	0.301 (92.1)	0.289	0.514	15.4	0.044 (3.5)	0.036 (1.4)	0.031 (1.1)	0.033 (1.5)	0.068 (1.6)	0.495 (95.1)	0.421	0.668	23.6
Ethalfuralin	0.044 (1.9)	0.036 (1.0)	0.031 (1.4)	0.040 (0.6)	0.065 (2.6)	0.317 (89.5)	0.287	0.548	16.4	0.044 (1.9)	0.036 (1.0)	0.031 (1.4)	0.051 (1.9)	0.060 (3.5)	0.452 (94.1)	0.384	0.662	20.8
Ethiofencarb	0.044 (2.0)	0.036 (0.9)	0.031 (1.3)	0.043 (0.2)	0.048 (2.2)	0.245 (90.4)	0.269	0.621	15.9	0.044 (2.0)	0.036 (0.9)	0.031 (1.3)	0.050 (1.7)	0.071 (3.4)	0.413 (92.1)	0.382	0.775	19.6
Ethofumasate	0.044 (3.4)	0.036 (1.2)	0.031 (0.9)	0.054 (0.4)	0.062 (2.1)	0.298 (88.7)	0.278	0.514	17.6	0.044 (3.4)	0.036 (1.2)	0.031 (0.9)	0.034 (1.2)	0.062 (2.6)	0.402 (90.8)	0.381	0.771	18.9
Ethoprophos	0.044 (3.3)	0.036 (1.1)	0.031 (1.1)	0.071 (0.3)	0.039 (1.8)	0.291 (93.1)	0.259	0.665	18.6	0.044 (3.3)	0.036 (1.1)	0.031 (1.1)	0.076 (2.2)	0.045 (1.6)	0.350 (88.4)	0.504	0.770	17.6
Etofenprox	0.044 (3.2)	0.036 (1.5)	0.031 (1.4)	0.023 (0.7)	0.065 (2.6)	0.298 (89.7)	0.287	0.541	17.8	0.044 (3.2)	0.036 (1.5)	0.031 (1.4)	0.071 (4.4)	0.061 (3.2)	0.318 (87.8)	0.501	0.776	15.9
Etoxazole	0.044 (3.1)	0.036 (1.6)	0.031 (1.3)	0.072 (4.3)	0.048 (2.2)	0.337 (92.6)	0.298	0.526	17.6	0.044 (3.1)	0.036 (1.6)	0.031 (1.3)	0.097 (6.3)	0.070 (3.3)	0.333 (92.1)	0.359	0.778	20.1
Famoxadone	0.044 (2.1)	0.036 (1.7)	0.031 (0.9)	0.008 (4.2)	0.048 (2.2)	0.298 (89.7)	0.301	0.668	15.9	0.044 (2.1)	0.036 (1.7)	0.031 (0.9)	0.096 (5.4)	0.064 (2.8)	0.368 (91.2)	0.385	0.776	21.8
Fenamidon	0.044 (3.2)	0.036 (1.4)	0.031 (0.7)	0.007 (4.1)	0.038 (1.8)	0.332 (85.6)	0.305	0.662	18.1	0.044 (3.2)	0.036 (1.4)	0.031 (0.7)	0.071 (2.1)	0.044 (1.7)	0.495 (95.1)	0.504	0.765	22.6
Fenamiphos (sum)	0.044 (3.1)	0.036 (1.5)	0.031 (0.8)	0.009 (3.9)	0.039 (1.8)	0.312 (92.4)	0.321	0.669	17.6	0.044 (3.1)	0.036 (1.5)	0.031 (0.8)	0.033 (1.5)	0.068 (1.6)	0.452 (94.1)	0.501	0.665	19.3
Fenarimol	0.044 (2.8)	0.036 (1.2)	0.031 (0.9)	0.070 (3.7)	0.041 (2.1)	0.298 (89.7)	0.315	0.662	15.9	0.044 (2.8)	0.036 (1.2)	0.031 (0.9)	0.051 (1.9)	0.060 (3.5)	0.413 (92.1)	0.328	0.654	18.9
Fenzaquin	0.044 (3.1)	0.036 (1.3)	0.031 (1.0)	0.066 (0.2)	0.068 (3.5)	0.301 (92.1)	0.311	0.665	16.5	0.044 (3.1)	0.036 (1.3)	0.031 (1.0)	0.076 (2.2)	0.071 (3.4)	0.402 (90.8)	0.329	0.774	15.6

Pesticide	Cucumber									Grapefruit								
	U <sub>i</sub> <sup>a</sup> (mg/kg (%))									U <sub>i</sub> <sup>a</sup> (mg/kg (%))								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)
(mg/kg)									(mg/kg)									
Fenbuconazole	0.044 (3.3)	0.036 (1.0)	0.031 (1.1)	0.052 (0.1)	0.039 (1.8)	0.317 (89.5)	0.314	0.514	15.3	0.044 (3.3)	0.036 (1.0)	0.031 (1.1)	0.071 (4.4)	0.062 (2.6)	0.350 (88.4)	0.385	0.771	16.2
Fenoxaprop-P-Ethyl	0.044 (2.3)	0.036 (0.9)	0.031 (1.4)	0.054 (0.6)	0.065 (2.6)	0.245 (90.4)	0.317	0.512	15.9	0.044 (2.3)	0.036 (0.9)	0.031 (1.4)	0.097 (6.3)	0.045 (1.6)	0.318 (87.8)	0.384	0.659	19.6
Fenoxycarb	0.044 (2.1)	0.036 (0.8)	0.031 (1.3)	0.051 (0.7)	0.048 (2.2)	0.298 (88.7)	0.298	0.628	14.8	0.044 (2.1)	0.036 (0.8)	0.031 (1.3)	0.096 (5.4)	0.061 (3.2)	0.333 (92.1)	0.394	0.662	20.3
Fenpropathrin	0.044 (2.0)	0.036 (1.6)	0.031 (0.9)	0.041 (0.5)	0.048 (2.2)	0.291 (93.1)	0.335	0.624	18.1	0.044 (2.0)	0.036 (1.6)	0.031 (0.9)	0.071 (2.1)	0.070 (3.3)	0.368 (91.2)	0.392	0.652	21.6
Fenpyroximate	0.044 (3.2)	0.036 (1.0)	0.031 (0.7)	0.040 (0.6)	0.038 (1.8)	0.298 (89.7)	0.289	0.701	16.9	0.044 (3.2)	0.036 (1.0)	0.031 (0.7)	0.033 (1.5)	0.064 (2.8)	0.495 (95.1)	0.389	1.014	18.9
Fenthion	0.044 (3.5)	0.036 (0.9)	0.031 (0.8)	0.043 (0.2)	0.039 (1.8)	0.285 (91.1)	0.287	0.704	15.8	0.044 (3.5)	0.036 (0.9)	0.031 (0.8)	0.051 (1.9)	0.044 (1.7)	0.452 (94.1)	0.374	0.665	17.8
Fenvalerate	0.044 (1.9)	0.036 (1.2)	0.031 (0.9)	0.054 (0.4)	0.041 (2.1)	0.331 (92.2)	0.269	0.517	17.5	0.044 (1.9)	0.036 (1.2)	0.031 (0.9)	0.076 (2.2)	0.068 (1.6)	0.413 (92.1)	0.365	1.011	16.9
Fipronil	0.044 (2.0)	0.036 (1.1)	0.031 (1.0)	0.071 (0.3)	0.068 (3.5)	0.337 (92.6)	0.278	0.514	15.8	0.044 (2.0)	0.036 (1.1)	0.031 (1.0)	0.071 (4.4)	0.060 (3.5)	0.402 (90.8)	0.361	0.774	20.1
Fluazifop-P-Butyl	0.044 (3.4)	0.036 (1.5)	0.031 (1.4)	0.041 (0.5)	0.062 (2.1)	0.298 (89.7)	0.259	0.548	16.4	0.044 (3.4)	0.036 (1.5)	0.031 (1.4)	0.097 (6.3)	0.071 (3.4)	0.350 (88.4)	0.368	0.776	23.4
Fluazinam	0.044 (3.3)	0.036 (1.6)	0.031 (1.3)	0.040 (0.6)	0.039 (1.8)	0.291 (93.1)	0.315	0.587	15.8	0.044 (3.3)	0.036 (1.6)	0.031 (1.3)	0.096 (5.4)	0.062 (2.6)	0.318 (87.8)	0.401	0.669	20.4
Fludioxynil	0.044 (3.2)	0.036 (1.7)	0.031 (0.9)	0.043 (0.2)	0.045 (1.8)	0.332 (85.6)	0.347	0.514	15.9	0.044 (3.2)	0.036 (1.7)	0.031 (0.9)	0.071 (2.1)	0.045 (1.6)	0.333 (92.1)	0.384	0.771	21.8
Flufenoxuron	0.044 (3.1)	0.036 (1.4)	0.031 (1.1)	0.054 (0.4)	0.048 (2.2)	0.312 (92.4)	0.325	0.665	18.1	0.044 (3.1)	0.036 (1.4)	0.031 (1.1)	0.033 (1.5)	0.061 (3.2)	0.368 (91.2)	0.382	0.778	19.8
Flurasulam	0.044 (2.1)	0.036 (1.5)	0.031 (1.4)	0.071 (0.3)	0.038 (1.8)	0.298 (89.7)	0.287	0.541	15.9	0.044 (2.1)	0.036 (1.5)	0.031 (1.4)	0.051 (1.9)	0.070 (3.3)	0.495 (95.1)	0.381	0.668	18.9
Flurochloridone	0.044 (3.2)	0.036 (1.2)	0.031 (1.3)	0.008 (4.2)	0.039 (1.8)	0.301 (92.1)	0.298	0.526	17.6	0.044 (3.2)	0.036 (1.2)	0.031 (1.3)	0.096 (5.4)	0.064 (2.8)	0.452 (94.1)	0.504	0.669	15.7
Flusilazole	0.044 (3.1)	0.036 (1.3)	0.031 (0.9)	0.007 (4.1)	0.041 (2.1)	0.317 (89.5)	0.301	0.668	18.6	0.044 (3.1)	0.036 (1.3)	0.031 (0.9)	0.071 (2.1)	0.044 (1.7)	0.413 (92.1)	0.501	0.770	16.4
Flutriafol	0.044 (2.8)	0.036 (1.0)	0.031 (0.7)	0.009 (3.9)	0.068 (3.5)	0.245 (90.4)	0.305	0.662	17.8	0.044 (2.8)	0.036 (1.0)	0.031 (0.7)	0.033 (1.5)	0.068 (1.6)	0.402 (90.8)	0.359	0.776	18.3
Foramsulfuron	0.044 (2.3)	0.036 (0.9)	0.031 (0.8)	0.070 (3.7)	0.039 (1.8)	0.298 (88.7)	0.321	0.669	17.6	0.044 (2.3)	0.036 (0.9)	0.031 (0.8)	0.051 (1.9)	0.060 (3.5)	0.350 (88.4)	0.385	0.778	19.5
Formatanate	0.044 (2.1)	0.036 (0.8)	0.031 (0.9)	0.066 (0.2)	0.065 (2.6)	0.285 (91.1)	0.315	0.665	15.9	0.044 (2.1)	0.036 (0.8)	0.031 (0.9)	0.076 (2.2)	0.071 (3.4)	0.318 (87.8)	0.504	0.776	15.6
Fosthiazate	0.044 (2.0)	0.036 (1.6)	0.031 (1.0)	0.052 (0.1)	0.048 (2.2)	0.337 (92.6)	0.311	0.663	18.1	0.044 (2.0)	0.036 (1.6)	0.031 (1.0)	0.071 (4.4)	0.062 (2.6)	0.333 (92.1)	0.501	0.765	16.2
Furathiocarb	0.044 (3.2)	0.036 (1.0)	0.031 (0.9)	0.054 (0.6)	0.048 (2.2)	0.298 (89.7)	0.314	0.664	17.6	0.044 (3.2)	0.036 (1.0)	0.031 (0.9)	0.097 (6.3)	0.045 (1.6)	0.368 (91.2)	0.328	0.998	19.6
Giberellic acid	0.044 (3.5)	0.036 (0.9)	0.031 (0.8)	0.051 (0.7)	0.038 (1.8)	0.332 (85.6)	0.317	0.521	15.9	0.044 (3.5)	0.036 (0.9)	0.031 (0.8)	0.096 (5.4)	0.061 (3.2)	0.495 (95.1)	0.329	0.884	20.3

Pesticide	Cucumber									Grapefruit										
	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))		U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup> (mg/kg)	U <sub>e</sub> (%)	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))		U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup> (mg/kg)	U <sub>e</sub> (%)
			U <sub>3</sub>	U <sub>3</sub>									U <sub>3</sub>	U <sub>3</sub>						
Halfenprox	0.044 (1.9)	0.036 (0.8)	0.031 (1.5)	0.041 (0.5)	0.039 (1.8)	0.312 (92.4)	0.298	0.547	18.4	0.044 (1.9)	0.036 (0.8)	0.031 (1.5)	0.071 (2.1)	0.070 (3.3)	0.452 (94.1)	0.385	0.771	21.6		
Haloxypop-2-Etoxyethyl	0.044 (2.0)	0.036 (1.6)	0.031 (1.6)	0.040 (0.6)	0.041 (2.1)	0.298 (89.7)	0.337	0.618	15.2	0.044 (2.0)	0.036 (1.6)	0.031 (1.6)	0.033 (1.5)	0.064 (2.8)	0.413 (92.1)	0.384	0.665	23.4		
Haloxypop-P-Methyl	0.044 (3.4)	0.036 (0.9)	0.031 (1.0)	0.043 (0.2)	0.068 (3.5)	0.301 (92.1)	0.289	0.662	16.5	0.044 (3.4)	0.036 (0.9)	0.031 (1.0)	0.051 (1.9)	0.044 (1.7)	0.402 (90.8)	0.394	0.882	21.8		
Hexaconazole	0.044 (3.3)	0.036 (1.2)	0.031 (1.1)	0.054 (0.4)	0.062 (2.1)	0.317 (89.5)	0.287	0.665	16.9	0.044 (3.3)	0.036 (1.2)	0.031 (1.1)	0.076 (2.2)	0.068 (1.6)	0.350 (88.4)	0.392	0.962	22.8		
Hexaflumuron	0.044 (3.2)	0.036 (1.1)	0.031 (1.4)	0.071 (0.3)	0.045 (1.8)	0.245 (90.4)	0.269	0.514	16.3	0.044 (3.2)	0.036 (1.1)	0.031 (1.4)	0.071 (4.4)	0.060 (3.5)	0.318 (87.8)	0.389	1.012	21.8		
Hexythiazox	0.044 (3.1)	0.036 (1.5)	0.031 (1.3)	0.041 (0.5)	0.039 (1.8)	0.298 (88.7)	0.278	0.512	16.1	0.044 (3.1)	0.036 (1.5)	0.031 (1.3)	0.097 (6.3)	0.071 (3.4)	0.333 (92.1)	0.374	0.669	23.6		
Imazalil	0.044 (2.1)	0.036 (1.6)	0.031 (0.9)	0.040 (0.6)	0.065 (2.6)	0.291 (93.1)	0.259	0.628	15.9	0.044 (2.1)	0.036 (1.6)	0.031 (0.9)	0.096 (5.4)	0.062 (2.6)	0.368 (91.2)	0.365	0.771	20.8		
Imazamox	0.044 (3.2)	0.036 (1.7)	0.031 (1.1)	0.043 (0.2)	0.048 (2.2)	0.285 (91.1)	0.339	0.624	17.5	0.044 (3.2)	0.036 (1.7)	0.031 (1.1)	0.071 (2.1)	0.045 (1.6)	0.495 (95.1)	0.361	0.778	19.6		
Imazapic	0.044 (3.1)	0.036 (1.4)	0.031 (1.4)	0.054 (0.4)	0.048 (2.2)	0.331 (92.2)	0.341	0.701	15.4	0.044 (3.1)	0.036 (1.4)	0.031 (1.4)	0.033 (1.5)	0.061 (3.2)	0.452 (94.1)	0.368	0.668	18.9		
Imazapyr	0.044 (2.8)	0.036 (1.5)	0.031 (1.3)	0.071 (0.3)	0.038 (1.8)	0.337 (92.6)	0.287	0.704	16.4	0.044 (2.8)	0.036 (1.5)	0.031 (1.3)	0.051 (1.9)	0.070 (3.3)	0.413 (92.1)	0.382	0.669	17.6		
Imazethapyr	0.044 (2.7)	0.036 (1.2)	0.031 (0.9)	0.072 (4.3)	0.039 (1.8)	0.298 (89.7)	0.298	0.517	15.7	0.044 (2.7)	0.036 (1.2)	0.031 (0.9)	0.097 (6.3)	0.064 (2.8)	0.402 (90.8)	0.381	0.770	15.9		
Imidacloprid	0.044 (2.5)	0.036 (1.3)	0.031 (0.7)	0.008 (4.2)	0.041 (2.1)	0.291 (93.1)	0.301	0.514	15.9	0.044 (2.5)	0.036 (1.3)	0.031 (0.7)	0.096 (5.4)	0.044 (1.7)	0.350 (88.4)	0.504	0.776	19.8		
Iodosulfuron-Methyl	0.044 (2.3)	0.036 (1.0)	0.031 (0.8)	0.007 (4.1)	0.068 (3.5)	0.332 (85.6)	0.305	0.548	17.6	0.044 (2.3)	0.036 (1.0)	0.031 (0.8)	0.071 (2.1)	0.068 (1.6)	0.318 (87.8)	0.501	0.778	18.6		
Ioxynil	0.044 (2.1)	0.036 (1.3)	0.031 (0.9)	0.009 (3.9)	0.065 (2.6)	0.312 (92.4)	0.321	0.587	18.6	0.044 (2.1)	0.036 (1.3)	0.031 (0.9)	0.033 (1.5)	0.060 (3.5)	0.333 (92.1)	0.359	0.776	23.6		
Iprodione	0.044 (2.0)	0.036 (1.0)	0.031 (1.0)	0.070 (3.7)	0.048 (2.2)	0.298 (89.7)	0.315	0.514	17.8	0.044 (2.0)	0.036 (1.0)	0.031 (1.0)	0.051 (1.9)	0.071 (3.4)	0.368 (91.2)	0.385	0.765	20.8		
Isoxaflutole	0.044 (3.2)	0.036 (0.9)	0.031 (1.1)	0.066 (0.2)	0.039 (1.8)	0.301 (92.1)	0.311	0.665	17.6	0.044 (3.2)	0.036 (0.9)	0.031 (1.1)	0.076 (2.2)	0.062 (2.6)	0.495 (95.1)	0.504	0.765	19.6		
Kresoxim methyl	0.044 (3.5)	0.036 (1.3)	0.031 (1.4)	0.052 (0.1)	0.052 (3.4)	0.317 (89.5)	0.314	0.541	15.9	0.044 (3.5)	0.036 (1.3)	0.031 (1.4)	0.071 (4.4)	0.045 (1.6)	0.452 (94.1)	0.501	0.665	18.9		
Lambda Cyhalothrin	0.044 (1.9)	0.036 (1.0)	0.031 (1.3)	0.054 (0.6)	0.037 (1.7)	0.245 (90.4)	0.317	0.526	18.1	0.044 (1.9)	0.036 (1.0)	0.031 (1.3)	0.097 (6.3)	0.061 (3.2)	0.413 (92.1)	0.328	0.654	17.6		
Lenacil	0.044 (2.0)	0.036 (0.9)	0.031 (0.9)	0.051 (0.7)	0.045 (1.8)	0.298 (88.7)	0.298	0.668	17.6	0.044 (2.0)	0.036 (0.9)	0.031 (0.9)	0.096 (5.4)	0.070 (3.3)	0.402 (90.8)	0.329	0.774	15.9		
Lufenuron	0.044 (3.4)	0.036 (0.8)	0.031 (0.7)	0.041 (0.5)	0.062 (2.1)	0.291 (93.1)	0.324	0.662	15.9	0.044 (3.4)	0.036 (0.8)	0.031 (0.7)	0.071 (2.1)	0.064 (2.8)	0.350 (88.4)	0.385	0.771	19.8		

Pesticide	Cucumber									Grapefruit								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (% )	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (% )
			U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>							U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>				
Malathion	0.044 (3.3)	0.036 (1.6)	0.031 (0.8)	0.040 (0.6)	0.048 (2.2)	0.337 (92.6)	0.289	0.669	14.6	0.044 (3.3)	0.036 (1.6)	0.031 (0.8)	0.033 (1.5)	0.044 (1.7)	0.318 (87.8)	0.384	0.659	20.1
MCPA	0.044 (3.2)	0.036 (1.2)	0.031 (0.9)	0.043 (0.2)	0.038 (1.8)	0.285 (91.1)	0.287	0.662	15.6	0.044 (3.2)	0.036 (1.2)	0.031 (0.9)	0.051 (1.9)	0.068 (1.6)	0.333 (92.1)	0.394	0.662	20.8
Mecoprop	0.044 (3.1)	0.036 (1.1)	0.031 (1.0)	0.054 (0.4)	0.039 (1.8)	0.329 (93.3)	0.269	0.547	12.3	0.044 (3.1)	0.036 (1.1)	0.031 (1.0)	0.076 (2.2)	0.060 (3.5)	0.368 (91.2)	0.392	0.652	22.8
Mefenpyr-diethyl	0.044 (2.1)	0.036 (1.5)	0.031 (1.0)	0.071 (0.3)	0.041 (2.1)	0.331 (92.2)	0.278	0.521	14.8	0.044 (2.1)	0.036 (1.5)	0.031 (1.0)	0.071 (4.4)	0.071 (3.4)	0.495 (95.1)	0.389	1.014	22.7
Mesosulfuron methyl	0.044 (3.2)	0.036 (1.6)	0.031 (1.1)	0.041 (0.5)	0.068 (3.5)	0.285 (91.1)	0.259	0.562	15.4	0.044 (3.2)	0.036 (1.6)	0.031 (1.1)	0.097 (6.3)	0.062 (2.6)	0.452 (94.1)	0.374	0.665	21.6
Mesotrione	0.044 (3.1)	0.036 (1.7)	0.031 (1.4)	0.040 (0.6)	0.039 (1.8)	0.337 (92.6)	0.321	0.574	14.6	0.044 (3.1)	0.036 (1.7)	0.031 (1.4)	0.096 (5.4)	0.045 (1.6)	0.413 (92.1)	0.365	1.011	20.4
Metalaxyl	0.044 (2.8)	0.036 (1.4)	0.031 (1.3)	0.043 (0.2)	0.052 (3.4)	0.298 (89.7)	0.287	0.618	14.5	0.044 (2.8)	0.036 (1.4)	0.031 (1.3)	0.071 (2.1)	0.061 (3.2)	0.402 (90.8)	0.361	0.774	19.8
Metamitron	0.044 (2.1)	0.036 (1.5)	0.031 (0.9)	0.054 (0.4)	0.037 (1.7)	0.332 (85.6)	0.298	0.662	17.2	0.044 (2.1)	0.036 (1.5)	0.031 (0.9)	0.033 (1.5)	0.070 (3.3)	0.350 (88.4)	0.368	0.776	15.7
Metconazole	0.044 (2.0)	0.036 (1.2)	0.031 (1.1)	0.071 (0.3)	0.045 (1.8)	0.312 (92.4)	0.301	0.665	17.1	0.044 (2.0)	0.036 (1.2)	0.031 (1.1)	0.051 (1.9)	0.064 (2.8)	0.318 (87.8)	0.501	0.669	16.4
Methidathion	0.044 (3.2)	0.036 (1.3)	0.031 (1.4)	0.008 (4.2)	0.048 (2.2)	0.298 (89.7)	0.305	0.514	15.6	0.044 (3.2)	0.036 (1.3)	0.031 (1.4)	0.096 (5.4)	0.044 (1.7)	0.333 (92.1)	0.425	0.771	18.3
Methomyl	0.044 (3.5)	0.036 (1.0)	0.031 (1.3)	0.007 (4.1)	0.038 (1.8)	0.301 (92.1)	0.321	0.512	18.1	0.044 (3.5)	0.036 (1.0)	0.031 (1.3)	0.071 (2.1)	0.068 (1.6)	0.368 (91.2)	0.384	0.778	19.5
Methoxyfenozide	0.044 (1.9)	0.036 (1.3)	0.031 (0.9)	0.009 (3.9)	0.039 (1.8)	0.317 (89.5)	0.315	0.628	16.4	0.044 (1.9)	0.036 (1.3)	0.031 (0.9)	0.033 (1.5)	0.060 (3.5)	0.495 (95.1)	0.382	0.668	15.6
Metolachlor	0.044 (2.2)	0.036 (1.0)	0.031 (0.7)	0.070 (3.7)	0.041 (2.1)	0.245 (90.4)	0.311	0.624	15.3	0.044 (2.2)	0.036 (1.0)	0.031 (0.7)	0.051 (1.9)	0.071 (3.4)	0.452 (94.1)	0.381	0.669	16.2
Metribuzin	0.044 (2.0)	0.036 (0.9)	0.031 (0.8)	0.066 (0.2)	0.068 (3.5)	0.298 (88.7)	0.314	0.701	15.8	0.044 (2.0)	0.036 (0.9)	0.031 (0.8)	0.076 (2.2)	0.062 (2.6)	0.413 (92.1)	0.504	0.770	19.6
Metsulfuron-methyl	0.044 (3.4)	0.036 (0.8)	0.031 (0.9)	0.052 (0.1)	0.065 (2.6)	0.291 (93.1)	0.317	0.704	15.9	0.044 (3.4)	0.036 (0.8)	0.031 (0.9)	0.071 (4.4)	0.045 (1.6)	0.402 (90.8)	0.501	0.776	20.3
Monocrotophos	0.044 (3.3)	0.036 (1.6)	0.031 (1.0)	0.054 (0.6)	0.048 (2.2)	0.285 (91.1)	0.298	0.517	17.6	0.044 (3.3)	0.036 (1.6)	0.031 (1.0)	0.097 (6.3)	0.061 (3.2)	0.350 (88.4)	0.359	0.778	21.6
Monolinuron	0.044 (3.2)	0.036 (0.9)	0.031 (1.1)	0.051 (0.7)	0.039 (1.8)	0.337 (92.6)	0.297	0.514	18.6	0.044 (3.2)	0.036 (0.9)	0.031 (1.1)	0.096 (5.4)	0.070 (3.3)	0.318 (87.8)	0.385	0.776	23.4
Myclobutanil	0.044 (3.1)	0.036 (1.2)	0.031 (1.4)	0.041 (0.5)	0.062 (2.1)	0.298 (89.7)	0.299	0.548	17.8	0.044 (3.1)	0.036 (1.2)	0.031 (1.4)	0.071 (2.1)	0.064 (2.8)	0.333 (92.1)	0.504	0.765	21.8
Nicosulfuron	0.044 (2.1)	0.036 (1.1)	0.031 (1.3)	0.040 (0.6)	0.045 (1.8)	0.332 (85.6)	0.289	0.514	17.6	0.044 (2.1)	0.036 (1.1)	0.031 (1.3)	0.033 (1.5)	0.044 (1.7)	0.368 (91.2)	0.501	1.011	22.8
Novaluron	0.044 (2.3)	0.036 (1.5)	0.031 (0.9)	0.043 (0.2)	0.048 (2.2)	0.312 (92.4)	0.287	0.665	15.9	0.044 (2.3)	0.036 (1.5)	0.031 (0.9)	0.051 (1.9)	0.068 (1.6)	0.495 (95.1)	0.328	0.994	21.8

Pesticide	Cucumber									Grapefruit								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (% )	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (% )
			U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>							U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>				
(mg/kg)									(mg/kg)									
Omethoate	0.044 (2.1)	0.036 (1.6)	0.031 (0.7)	0.054 (0.4)	0.038 (1.8)	0.298 (89.7)	0.269	0.541	18.1	0.044 (2.1)	0.036 (1.6)	0.031 (0.7)	0.076 (2.2)	0.060 (3.5)	0.452 (94.1)	0.329	0.886	23.6
Oxadixyl	0.044 (1.9)	0.036 (1.7)	0.031 (0.8)	0.071 (0.3)	0.039 (1.8)	0.301 (92.1)	0.278	0.526	17.6	0.044 (1.9)	0.036 (1.7)	0.031 (0.8)	0.071 (4.4)	0.071 (3.4)	0.413 (92.1)	0.385	0.887	20.8
Oxamyl	0.044 (3.2)	0.036 (1.4)	0.031 (0.9)	0.041 (0.5)	0.041 (2.1)	0.317 (89.5)	0.259	0.668	15.9	0.044 (3.2)	0.036 (1.4)	0.031 (0.9)	0.097 (6.3)	0.062 (2.6)	0.402 (90.8)	0.384	0.885	19.6
Oxyflourfen	0.044 (3.1)	0.036 (1.5)	0.031 (1.0)	0.040 (0.6)	0.068 (3.5)	0.245 (90.4)	0.317	0.662	16.3	0.044 (3.1)	0.036 (1.5)	0.031 (1.0)	0.096 (5.4)	0.045 (1.6)	0.350 (88.4)	0.394	0.881	18.9
Parathion Methyl	0.044 (2.8)	0.036 (1.2)	0.031 (1.4)	0.043 (0.2)	0.052 (3.4)	0.298 (88.7)	0.287	0.669	15.6	0.044 (2.8)	0.036 (1.2)	0.031 (1.4)	0.071 (2.1)	0.061 (3.2)	0.318 (87.8)	0.392	0.669	17.6
Penconazole	0.044 (2.1)	0.036 (1.3)	0.031 (1.3)	0.054 (0.4)	0.037 (1.7)	0.332 (85.6)	0.298	0.618	16.3	0.044 (2.1)	0.036 (1.3)	0.031 (1.3)	0.033 (1.5)	0.070 (3.3)	0.333 (92.1)	0.389	0.771	15.9
Pendimethalin	0.044 (2.0)	0.036 (1.0)	0.031 (0.9)	0.071 (0.3)	0.045 (1.8)	0.312 (92.4)	0.301	0.662	15.6	0.044 (2.0)	0.036 (1.0)	0.031 (0.9)	0.051 (1.9)	0.064 (2.8)	0.368 (91.2)	0.374	0.778	18.3
Permethrin	0.044 (3.2)	0.036 (1.0)	0.031 (1.1)	0.023 (0.7)	0.065 (2.6)	0.298 (89.7)	0.305	0.665	18.1	0.044 (3.2)	0.036 (1.0)	0.031 (1.1)	0.096 (5.4)	0.044 (1.7)	0.495 (95.1)	0.365	0.668	17.5
Phenmedipham	0.044 (3.5)	0.036 (0.9)	0.031 (1.4)	0.072 (4.3)	0.048 (2.2)	0.291 (93.1)	0.321	0.514	14.5	0.044 (3.5)	0.036 (0.9)	0.031 (1.4)	0.071 (2.1)	0.068 (1.6)	0.452 (94.1)	0.361	0.669	17.3
Phenthoate	0.044 (1.9)	0.036 (1.2)	0.031 (1.3)	0.008 (4.2)	0.062 (2.1)	0.285 (91.1)	0.315	0.512	16.2	0.044 (1.9)	0.036 (1.2)	0.031 (1.3)	0.033 (1.5)	0.060 (3.5)	0.413 (92.1)	0.368	0.770	17.2
Phosalone	0.044 (2.0)	0.036 (1.1)	0.031 (0.9)	0.007 (4.1)	0.039 (1.8)	0.329 (93.3)	0.311	0.628	18.2	0.044 (2.0)	0.036 (1.1)	0.031 (0.9)	0.051 (1.9)	0.071 (3.4)	0.402 (90.8)	0.441	0.776	19.6
Phosmet	0.044 (3.4)	0.036 (1.5)	0.031 (0.7)	0.009 (3.9)	0.065 (2.6)	0.337 (92.6)	0.314	0.624	14.3	0.044 (3.4)	0.036 (1.5)	0.031 (0.7)	0.076 (2.2)	0.062 (2.6)	0.350 (88.4)	0.445	0.778	15.6
Pirimicarb	0.044 (3.3)	0.036 (1.6)	0.031 (0.8)	0.070 (3.7)	0.048 (2.2)	0.298 (89.7)	0.317	0.701	16.9	0.044 (3.3)	0.036 (1.6)	0.031 (0.8)	0.071 (4.4)	0.045 (1.6)	0.318 (87.8)	0.384	0.776	16.2
Pirimiphos methyl	0.044 (3.2)	0.036 (1.7)	0.031 (0.9)	0.066 (0.2)	0.048 (2.2)	0.301 (92.1)	0.298	0.704	14.8	0.044 (3.2)	0.036 (1.7)	0.031 (0.9)	0.097 (6.3)	0.061 (3.2)	0.333 (92.1)	0.382	0.765	19.6
Prochloraz	0.044 (3.1)	0.036 (1.4)	0.031 (1.0)	0.052 (0.1)	0.038 (1.8)	0.317 (89.5)	0.315	0.517	15.9	0.044 (3.1)	0.036 (1.4)	0.031 (1.0)	0.096 (5.4)	0.070 (3.3)	0.368 (91.2)	0.381	0.765	20.3
Procymidone	0.044 (2.1)	0.036 (1.5)	0.031 (0.8)	0.054 (0.6)	0.039 (1.8)	0.245 (90.4)	0.347	0.514	17.6	0.044 (2.1)	0.036 (1.5)	0.031 (0.8)	0.071 (2.1)	0.064 (2.8)	0.495 (95.1)	0.504	0.665	21.6
Profenofos	0.044 (2.3)	0.036 (1.2)	0.031 (1.5)	0.051 (0.7)	0.041 (2.1)	0.298 (88.7)	0.325	0.548	18.6	0.044 (2.3)	0.036 (1.2)	0.031 (1.5)	0.033 (1.5)	0.044 (1.7)	0.452 (94.1)	0.501	0.654	20.8
Profoxydim (Clefoxydim)	0.044 (2.1)	0.036 (1.3)	0.031 (1.6)	0.041 (0.5)	0.068 (3.5)	0.332 (85.6)	0.298	0.587	17.8	0.044 (2.1)	0.036 (1.3)	0.031 (1.6)	0.051 (1.9)	0.068 (1.6)	0.413 (92.1)	0.359	0.774	19.6
Prometryn	0.044 (1.9)	0.036 (1.0)	0.031 (1.0)	0.040 (0.6)	0.045 (1.7)	0.312 (92.4)	0.289	0.514	17.6	0.044 (1.9)	0.036 (1.0)	0.031 (1.0)	0.076 (2.2)	0.060 (3.5)	0.402 (90.8)	0.385	0.771	17.6
Propaquizafop	0.044 (3.2)	0.036 (1.3)	0.031 (1.1)	0.043 (0.2)	0.039 (1.8)	0.298 (89.7)	0.287	0.665	15.9	0.044 (3.2)	0.036 (1.3)	0.031 (1.1)	0.071 (4.4)	0.071 (3.4)	0.350 (88.4)	0.504	0.659	18.2
Propargite	0.044 (3.1)	0.036 (1.0)	0.031 (1.4)	0.054 (0.4)	0.052 (3.4)	0.291 (93.1)	0.269	0.541	18.1	0.044 (3.1)	0.036 (1.0)	0.031 (1.4)	0.097 (6.3)	0.062 (2.6)	0.318 (87.8)	0.501	0.662	17.1

Pesticide	Cucumber									Grapefruit								
	U <sub>i</sub> <sup>a</sup> (mg/kg (%))									U <sub>i</sub> <sup>a</sup> (mg/kg (%))								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)
(mg/kg)									(mg/kg)									
Propazine	0.044 (2.8)	0.036 (0.9)	0.031 (1.3)	0.071 (0.3)	0.037 (1.7)	0.285 (91.1)	0.278	0.526	17.6	0.044 (2.8)	0.036 (0.9)	0.031 (1.3)	0.096 (5.4)	0.045 (1.6)	0.333 (92.1)	0.328	0.652	18.7
Propoxycarbazon	0.044 (2.1)	0.036 (0.8)	0.031 (0.9)	0.041 (0.5)	0.045 (1.8)	0.337 (92.6)	0.259	0.668	15.9	0.044 (2.1)	0.036 (0.8)	0.031 (0.9)	0.071 (2.1)	0.061 (3.2)	0.368 (91.2)	0.329	1.014	18.8
Propyconazole	0.044 (2.0)	0.036 (1.6)	0.031 (1.1)	0.040 (0.6)	0.048 (2.2)	0.298 (89.7)	0.315	0.662	18.5	0.044 (2.0)	0.036 (1.6)	0.031 (1.1)	0.033 (1.5)	0.070 (3.3)	0.495 (95.1)	0.385	0.665	16.4
Propyzamide	0.044 (3.2)	0.036 (1.2)	0.031 (1.4)	0.043 (0.2)	0.038 (1.8)	0.301 (92.1)	0.347	0.669	14.5	0.044 (3.2)	0.036 (1.2)	0.031 (1.4)	0.051 (1.9)	0.064 (2.8)	0.452 (94.1)	0.384	1.011	18.3
Prothopos	0.044 (3.5)	0.036 (1.1)	0.031 (1.3)	0.054 (0.4)	0.039 (1.8)	0.317 (89.5)	0.325	0.514	16.9	0.044 (3.5)	0.036 (1.1)	0.031 (1.3)	0.071 (4.4)	0.044 (1.7)	0.413 (92.1)	0.394	0.774	19.5
Pymetrozine	0.044 (1.9)	0.036 (1.5)	0.031 (0.9)	0.071 (0.3)	0.041 (2.1)	0.245 (90.4)	0.317	0.618	15.6	0.044 (1.9)	0.036 (1.5)	0.031 (0.9)	0.097 (6.3)	0.068 (1.6)	0.402 (90.8)	0.392	0.776	15.6
Pyraclostrobin	0.044 (2.0)	0.036 (1.6)	0.031 (0.7)	0.008 (4.2)	0.068 (3.5)	0.298 (88.7)	0.315	0.662	18.2	0.044 (2.0)	0.036 (1.6)	0.031 (0.7)	0.096 (5.4)	0.060 (3.5)	0.350 (88.4)	0.389	0.669	16.2
Pyraflufen-Ethyl	0.044 (3.4)	0.036 (1.7)	0.031 (0.8)	0.007 (4.1)	0.052 (3.4)	0.332 (85.6)	0.347	0.665	17.6	0.044 (3.4)	0.036 (1.7)	0.031 (0.8)	0.071 (2.1)	0.071 (3.4)	0.318 (87.8)	0.374	0.771	19.6
Pyrazophos	0.044 (3.3)	0.036 (1.4)	0.031 (0.9)	0.009 (3.9)	0.037 (1.7)	0.312 (92.4)	0.325	0.514	13.6	0.044 (3.3)	0.036 (1.4)	0.031 (0.9)	0.033 (1.5)	0.062 (2.6)	0.333 (92.1)	0.365	0.778	20.3
Pyridaben	0.044 (3.2)	0.036 (1.5)	0.031 (1.0)	0.070 (3.7)	0.045 (1.8)	0.298 (89.7)	0.287	0.512	15.2	0.044 (3.2)	0.036 (1.5)	0.031 (1.0)	0.051 (1.9)	0.045 (1.6)	0.368 (91.2)	0.361	0.668	21.6
Pyridaphention	0.044 (3.1)	0.036 (1.2)	0.031 (1.1)	0.066 (0.2)	0.065 (2.8)	0.291 (93.1)	0.298	0.628	14.6	0.044 (3.1)	0.036 (1.2)	0.031 (1.1)	0.076 (2.2)	0.061 (3.2)	0.495 (95.1)	0.368	0.669	23.4
Pyridate	0.044 (2.1)	0.036 (1.3)	0.031 (1.4)	0.052 (0.1)	0.039 (1.8)	0.285 (91.1)	0.301	0.624	12.5	0.044 (2.1)	0.036 (1.3)	0.031 (1.4)	0.071 (4.4)	0.070 (3.3)	0.452 (94.1)	0.384	0.770	21.8
Pyriproxyfen	0.044 (2.3)	0.036 (1.0)	0.031 (1.3)	0.054 (0.6)	0.065 (2.6)	0.337 (92.6)	0.305	0.701	13.6	0.044 (2.3)	0.036 (1.0)	0.031 (1.3)	0.097 (6.3)	0.064 (2.8)	0.413 (92.1)	0.382	0.776	22.8
Quizalofop-P-Ethyl	0.044 (2.1)	0.036 (1.3)	0.031 (0.9)	0.051 (0.7)	0.048 (2.2)	0.298 (89.7)	0.321	0.704	14.8	0.044 (2.1)	0.036 (1.3)	0.031 (0.9)	0.096 (5.4)	0.044 (1.7)	0.402 (90.8)	0.381	0.778	21.8
Rimsulfuron	0.044 (1.9)	0.036 (1.0)	0.031 (0.7)	0.041 (0.5)	0.048 (2.2)	0.301 (92.1)	0.315	0.517	15.9	0.044 (1.9)	0.036 (1.0)	0.031 (0.7)	0.071 (2.1)	0.068 (1.6)	0.350 (88.4)	0.504	0.776	23.6
Sethoxydim	0.044 (3.2)	0.036 (0.9)	0.031 (0.8)	0.040 (0.6)	0.038 (1.8)	0.317 (89.5)	0.311	0.514	17.6	0.044 (3.2)	0.036 (0.9)	0.031 (0.8)	0.033 (1.5)	0.060 (3.5)	0.318 (87.8)	0.501	0.765	20.8
Simozine	0.044 (3.1)	0.036 (0.8)	0.031 (0.9)	0.043 (0.2)	0.039 (1.8)	0.245 (90.4)	0.314	0.548	18.6	0.044 (3.1)	0.036 (0.8)	0.031 (0.9)	0.051 (1.9)	0.071 (3.4)	0.333 (92.1)	0.359	0.884	19.6
Spirodiclofen	0.044 (2.8)	0.036 (1.6)	0.031 (1.0)	0.054 (0.4)	0.041 (2.1)	0.298 (88.7)	0.317	0.514	15.6	0.044 (2.8)	0.036 (1.6)	0.031 (1.0)	0.076 (2.2)	0.062 (2.6)	0.368 (91.2)	0.385	0.765	18.9
Spiroxamine	0.044 (2.3)	0.036 (1.1)	0.031 (0.8)	0.071 (0.3)	0.068 (3.5)	0.332 (85.6)	0.298	0.665	15.2	0.044 (2.3)	0.036 (1.1)	0.031 (0.8)	0.071 (4.4)	0.045 (1.6)	0.495 (95.1)	0.504	0.665	17.6
Sulfosulfuron	0.044 (2.1)	0.036 (1.5)	0.031 (1.5)	0.041 (0.5)	0.070 (3.4)	0.312 (92.4)	0.315	0.541	17.8	0.044 (2.1)	0.036 (1.5)	0.031 (1.5)	0.097 (6.3)	0.061 (3.2)	0.452 (94.1)	0.501	0.654	15.9
Tau-Fluvalinate	0.044 (2.0)	0.036 (1.6)	0.031 (1.6)	0.040 (0.6)	0.039 (1.8)	0.298 (89.7)	0.347	0.526	17.9	0.044 (2.0)	0.036 (1.6)	0.031 (1.6)	0.096 (5.4)	0.070 (3.3)	0.413 (92.1)	0.328	0.774	20.6

Pesticide	Cucumber									Grapefruit									
	U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)		U <sub>1</sub>	U <sub>2</sub>	U <sub>i</sub> <sup>a</sup> (mg/kg (%))			U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>e</sub> (%)
			U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>								U <sub>3</sub>	U <sub>4</sub>	U <sub>5</sub>				
(mg/kg)									(mg/kg)										
Tebuconazole	0.044 (3.2)	0.036 (1.7)	0.031 (1.0)	0.043 (0.2)	0.052 (3.4)	0.291 (93.1)	0.325	0.668	12.3	0.044 (3.2)	0.036 (1.7)	0.031 (1.0)	0.071 (2.1)	0.064 (2.8)	0.402 (90.8)	0.329	0.771	22.3	
Tebufenozide	0.044 (3.5)	0.036 (1.4)	0.031 (1.1)	0.054 (0.4)	0.037 (1.7)	0.337 (92.6)	0.314	0.662	15.6	0.044 (3.5)	0.036 (1.4)	0.031 (1.1)	0.033 (1.5)	0.044 (1.7)	0.350 (88.4)	0.385	0.659	22.9	
Teflubenzuron	0.044 (1.9)	0.036 (1.5)	0.031 (1.4)	0.071 (0.3)	0.045 (1.8)	0.298 (89.7)	0.289	0.669	14.3	0.044 (1.9)	0.036 (1.5)	0.031 (1.4)	0.051 (1.9)	0.068 (1.6)	0.318 (87.8)	0.384	0.662	21.8	
Tepraloxymid	0.044 (2.0)	0.036 (1.2)	0.031 (1.3)	0.041 (0.5)	0.048 (2.2)	0.301 (92.1)	0.287	0.662	15.6	0.044 (2.0)	0.036 (1.2)	0.031 (1.3)	0.076 (2.2)	0.060 (3.5)	0.333 (92.1)	0.394	0.652	15.6	
Terbutylazine	0.044 (3.4)	0.036 (1.3)	0.031 (0.9)	0.040 (0.6)	0.038 (1.8)	0.317 (89.5)	0.269	0.665	18.5	0.044 (3.4)	0.036 (1.3)	0.031 (0.9)	0.071 (4.4)	0.071 (3.4)	0.368 (91.2)	0.392	1.014	16.2	
Terbutryn	0.044 (3.3)	0.036 (1.0)	0.031 (0.7)	0.043 (0.2)	0.039 (1.8)	0.245 (90.4)	0.278	0.514	14.3	0.044 (3.3)	0.036 (1.0)	0.031 (0.7)	0.097 (6.3)	0.062 (2.6)	0.495 (95.1)	0.389	0.665	19.6	
Tetraconazole	0.044 (3.2)	0.036 (1.3)	0.031 (0.8)	0.054 (0.4)	0.041 (2.1)	0.298 (88.7)	0.259	0.512	15.2	0.044 (3.2)	0.036 (1.3)	0.031 (0.8)	0.096 (5.4)	0.045 (1.6)	0.452 (94.1)	0.374	1.011	20.3	
Thiabendazole	0.044 (3.1)	0.036 (1.0)	0.031 (0.9)	0.071 (0.3)	0.068 (3.5)	0.289 (88.9)	0.311	0.628	15.6	0.044 (3.1)	0.036 (1.0)	0.031 (0.9)	0.071 (2.1)	0.061 (3.2)	0.413 (92.1)	0.365	0.774	21.6	
Thiacloprid	0.044 (2.1)	0.036 (0.9)	0.031 (1.0)	0.007 (4.1)	0.052 (3.4)	0.288 (96.5)	0.287	0.624	18.1	0.044 (2.1)	0.036 (0.9)	0.031 (1.0)	0.033 (1.5)	0.070 (3.3)	0.402 (90.8)	0.361	0.776	19.5	
Thiamethoxam	0.044 (2.3)	0.036 (0.8)	0.031 (1.1)	0.009 (3.9)	0.037 (1.7)	0.291 (93.1)	0.298	0.701	14.5	0.044 (2.3)	0.036 (0.8)	0.031 (1.1)	0.051 (1.9)	0.064 (2.8)	0.350 (88.4)	0.368	0.669	18.2	
Thifensulfuron-methyl	0.044 (2.1)	0.036 (1.6)	0.031 (1.4)	0.070 (3.7)	0.045 (1.8)	0.332 (85.6)	0.301	0.704	16.7	0.044 (2.1)	0.036 (1.6)	0.031 (1.4)	0.076 (2.2)	0.044 (1.7)	0.318 (87.8)	0.381	0.771	18.1	
Thiohanate methyl	0.044 (1.9)	0.036 (1.1)	0.031 (1.3)	0.066 (0.2)	0.072 (41)	0.312 (92.4)	0.305	0.517	16.9	0.044 (1.9)	0.036 (1.1)	0.031 (1.3)	0.071 (4.4)	0.068 (1.6)	0.333 (92.1)	0.504	0.778	17.3	
Thiometon	0.044 (3.2)	0.036 (1.5)	0.031 (0.9)	0.052 (0.1)	0.039 (1.8)	0.298 (89.7)	0.321	0.514	17.6	0.044 (3.2)	0.036 (1.5)	0.031 (0.9)	0.097 (6.3)	0.060 (3.5)	0.368 (91.2)	0.501	0.668	17.1	
Tolyfluanid	0.044 (3.1)	0.036 (1.6)	0.031 (0.7)	0.054 (0.6)	0.052 (3.4)	0.332 (94.6)	0.315	0.548	17.2	0.044 (3.1)	0.036 (1.6)	0.031 (0.7)	0.096 (5.4)	0.071 (3.4)	0.495 (95.1)	0.359	0.669	15.7	
Tralkoxydim	0.044 (2.8)	0.036 (1.7)	0.031 (0.8)	0.051 (0.7)	0.037 (1.7)	0.301 (92.1)	0.311	0.623	15.9	0.044 (2.8)	0.036 (1.7)	0.031 (0.8)	0.071 (2.1)	0.062 (2.6)	0.452 (94.1)	0.385	0.765	16.4	
Triadimefon	0.044 (2.4)	0.036 (1.4)	0.031 (0.9)	0.041 (0.5)	0.045 (1.8)	0.317 (89.5)	0.314	0.654	14.8	0.044 (2.4)	0.036 (1.4)	0.031 (0.9)	0.033 (1.5)	0.045 (1.6)	0.413 (92.1)	0.504	0.665	18.3	
Triadimenol	0.044 (3.1)	0.036 (1.5)	0.031 (0.8)	0.040 (0.6)	0.074 (3.8)	0.245 (90.4)	0.317	0.681	15.9	0.044 (3.1)	0.036 (1.5)	0.031 (0.8)	0.051 (1.9)	0.061 (3.2)	0.402 (90.8)	0.501	0.654	19.5	
Triallate	0.044 (2.3)	0.036 (1.2)	0.031 (1.5)	0.043 (0.2)	0.048 (2.2)	0.298 (88.7)	0.298	0.618	17.6	0.044 (2.3)	0.036 (1.2)	0.031 (1.5)	0.034 (1.2)	0.070 (3.3)	0.350 (88.4)	0.328	0.774	15.6	
Triasulfuron	0.044 (2.1)	0.036 (1.3)	0.031 (1.6)	0.054 (0.4)	0.038 (1.8)	0.335 (95.6)	0.301	0.662	18.6	0.044 (2.1)	0.036 (1.3)	0.031 (1.6)	0.076 (2.2)	0.064 (2.8)	0.318 (87.8)	0.329	0.771	16.2	
Tribenuron-Methyl	0.044 (2.0)	0.036 (1.0)	0.031 (1.0)	0.071 (0.3)	0.039 (1.8)	0.332 (85.6)	0.308	0.665	17.8	0.044 (2.0)	0.036 (1.0)	0.031 (1.0)	0.071 (4.4)	0.044 (1.7)	0.333 (92.1)	0.385	0.659	19.6	
Trichlorfon	0.044 (3.2)	0.036 (1.0)	0.031 (1.1)	0.008 (4.2)	0.041 (2.1)	0.312 (92.4)	0.296	0.514	17.6	0.044 (3.2)	0.036 (1.0)	0.031 (1.1)	0.097 (6.3)	0.068 (1.6)	0.368 (91.2)	0.384	0.662	20.3	

Pesticide	Cucumber									Grapefruit								
	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub> <sup>a</sup> (mg/kg (%))	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>c</sub> (% )	U <sub>1</sub>	U <sub>2</sub>	U <sub>3</sub> <sup>a</sup> (mg/kg (%))	U <sub>4</sub>	U <sub>5</sub>	U <sub>6</sub>	U <sub>c</sub>	U <sub>e</sub> <sup>b</sup>	U <sub>c</sub> (% )
Trifloxystrobin	0.044 (3.5)	0.036 (0.9)	0.031 (1.4)	0.007 (4.1)	0.068 (3.5)	0.298 (89.7)	0.289	0.512	15.9	0.044 (3.5)	0.036 (0.9)	0.031 (1.4)	0.096 (5.4)	0.060 (3.5)	0.495 (95.1)	0.394	0.652	21.6
Triflumizole	0.044 (1.9)	0.036 (1.2)	0.031 (1.3)	0.009 (3.9)	0.039 (1.8)	0.301 (92.1)	0.287	0.628	18.1	0.044 (1.9)	0.036 (1.2)	0.031 (1.3)	0.071 (2.1)	0.071 (3.4)	0.452 (94.1)	0.392	1.014	23.4
Triflumuron	0.044 (2.3)	0.036 (1.1)	0.031 (0.9)	0.070 (3.7)	0.052 (3.4)	0.317 (89.5)	0.269	0.624	17.6	0.044 (2.3)	0.036 (1.1)	0.031 (0.9)	0.033 (1.5)	0.062 (2.6)	0.413 (92.1)	0.389	0.665	21.8
Triticonazole	0.044 (2.1)	0.036 (1.5)	0.031 (1.5)	0.066 (0.2)	0.037 (1.7)	0.245 (90.4)	0.278	0.701	15.9	0.044 (2.1)	0.036 (1.5)	0.031 (1.5)	0.051 (1.9)	0.045 (1.6)	0.402 (90.8)	0.374	1.011	22.8
2.4 D Acid	0.044 (1.9)	0.036 (1.6)	0.031 (0.8)	0.052 (0.1)	0.045 (1.8)	0.298 (88.7)	0.259	0.704	18.7	0.044 (1.9)	0.036 (1.6)	0.031 (0.8)	0.071 (4.4)	0.061 (3.2)	0.452 (94.1)	0.365	0.774	21.8

U<sub>i</sub><sup>a</sup> are the contributions of each uncertainty source to U<sub>c</sub> of the method.

U<sub>e</sub><sup>b</sup> is the expanded uncertainty for a 4 mg/kg estimated for a level of confidence of 95%

## Conclusions

In this paper, an analytical method for the separation of 233 pesticide in cucumber and grapefruits using QuEChERS method and GC-MS, LC-MS/MS is presented. The validation data showed satisfactory performance of analytical method, in terms of specificity, sensitivity, linearity, recovery, precision and  $U_e$  according to the European SANTE/11945/2015 Guideline. One or multiple pesticide residues were detected in 11.6% of cucumber and 13.8% of grapefruit samples, but the levels were below the EU MRLs. The residues found in cucumbers and grapefruits were belonging to the group of fungicides, insecticides, herbicide, acaricide and plant growth regulator. The most frequently detected residues in cucumber samples were Clofentezine (4.4%), propyzamide (3.96), propamocarb (3.03), Azinphosethyl (1.8), metalxyl, procymidone, chlorpyrifos and Carbendazim. However, the fungicide propamocarb (8.8%) was the most common residue in cucumbers, followed by propyzamide (6.3%) and Clofentezine (2%). The fungicide fludioxonil was the most prominent pesticide found in grapefruit samples, with a frequency of 9.1%. Imazalil was the second most commonly found pesticide in grapefruits with 4.7% of the samples, followed by Thiabendazol, quinophos and carbaryl. The proposed method was also found to be suitable for different kinds of fruits.

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