

# Original article (Orijinal araştırma)

# Acute and chronic exposure risks of insecticide residues in fresh commodities collected from Bursa (Türkiye) province markets during winter season1

Kıs sezonunda Bursa ili (Türkiye) satıs noktalarından toplanan farklı taze tüketim ürünlerindeki insektisit kalıntılarının akut ve kronik risk değerlendirmesi

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#### **Abstract**

This study shows the findings about pesticide residues and the associated acute and chronic exposure risks of different fresh commodities collected from different markets located in Bursa province (Türkiye) during 2023 winter season. For this purpose, pesticide residue levels of the collected samples were analysed with LC-MS/MS. Highest levels of insecticide and acaricide residues were detected in some lettuce, parsley, dill, carrot, pear, mandarin and banana samples and they were exceeded the maximum residue limit (MRL). The acute and the chronic exposures to pesticides were assessed by using the highest and the average redidue levels of each pesticide respectively. Highest acute exposure was calculated as acute reference dose (ARfD) exceedance rate and it was 104.27% for indoxacarb in apples, 107.06% and 137.11% for lambda-cyhalothrin in pears and mandarins, and 158.2% for phosmet in pears. For all commodity types, none of the pesticide residues displayed chronic hazard. When the cumulative long-term exposure evaluated, none of the insecticides was found to be risky for adults. The findings showed that the levels of insecticide residues on lettuce, parsley, dill, carrot, apple, pear, mandarin, orange and banana samples collected from Bursa markets in winter 2023 could not be considered as an important public health risk.

Keywords: Acute, chronic, insecticide residues, risk assessment

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Bu çalışma, 2023 yılı kış sezonunda Bursa ili (Türkiye) yerel satış noktalarından toplanan farklı taze tüketim ürünleri üzerindeki pestisit kalıntıları ve bunların tüketiciler üzerine olan akut ve kronik maruziyet risklerine ait bulguları rapor etmektedir. Bu amacla toplanan örneklerin LC-MS/MS kullanılarak kalıntı düzevleri tespit edilmiştir. Bulgulara göre, toplanan bazı marul, maydanoz, dereotu, havuç, armut, mandalina ve muz örneklerinde tespit edilen en yüksek insektisit ve akarisit kalıntıları maksimum kalıntı limitlerini (MRL) aşmıştır. Akut ve kronik maruziyetler, pestisitlerin ortalama ve en yüksek kalıntı konsantrasyonları kullanılarak değerlendirilmiştir. En yüksek akut tehlike, akut referans doz asımı (ARfD) olarak hesaplanmıştır ve bu değer indoxacarb için elmada %104.27, lambda-cyhalothrin için armut ve mandalinada sırasıyla %107.06 ve %137.11 ve phosmet için armutta %158.2 olarak bulunmuştur. Tüm ürünlerde her bir pestisit kalıntısı için kronik tehlike gözlenmemiştir. Kümülatif uzun süreli maruz kalma değerlendirildiğinde, yetişkinler için hiçbir insektisitin risk oluşturmadığı tespit edilmiştir. Bulgular, 2023 yılında Bursa pazarlarından toplanan marul, maydanoz, dereotu, havuc, elma, armut, mandalina, portakal ve muz örneklerinde insektisit kalıntılarının görülmesinin büyük bir halk sağlığı riski olarak değerlendirilemeyeceğini göstermektedir.

Anahtar sözcükler: Akut, kronik, insektisit kalıntıları, risk değerlendirmesi

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## Introduction

Türkiye is one of the largest fruit and vegetable producer, following China, India, Brazil and the USA (FAO, 2021). According to the data from Turkish Statistical Institute, 19.5 million tons of fruits and 25.6 million tons of vegetables were produced in 2021 in Türkiye. Previous studies have showed that some vegetables and fruits have protective impacts against the development of serious human diseases such as cardiovascular problems, diabetes, obesity and cancer (Ferretti et al., 2010). Their protective roles could be originated from the various nutrients which contain fiber, vitamins and phytonutrients (Prior, 2003). For these reasons, health authorities encourage that consumers eat at least five portions of fresh fruit and vegetables daily (TÜBER, 2019). Besides health benefits of fruit and vegatables, the agricultural chemicals which are widely used to control pests during their cultivations may lead to health problems for consumers (Baldi et al., 2001; Lozowicka, 2015). Some insecticides have been related with a wide range of human health hazards, ranging from acute to chronic impacts (Calvert et al., 2001; Bhanti & Taneja, 2007). Chronic health effects (such as, various types of cancers, disorders in the endocrine, reproductive system, and embryonic development) may occur years after even minimal exposure to pesticides in the environment, food and water (Berrada et al., 2010; Yousefi et al., 2022). The long-term health problems are particularly serious when these commodities are consumed continuously as fresh and processed foods (Solecki et al., 2005).

In order to protect public health, regular monitoring of insecticide residues and dietary risk assessment are important tasks for human health. For this reason, based on the maximum residue limits (MRL) for each insecticide and commodity, their residues are regularly monitored in fresh foods as they are eaten raw (Ambrus et al., 2023). Nevertheless, the insecticide residues above their MRL may be detected on fresh fruits and vegetables. The reasons for the residues are (1) paying insufficient attention to pre-harvest Interval (PHI), (2) the use of very high dose of pesticides due to development of resistance in pests, (3) the use of pesticide mixtures in order to provide broad spectrum protection against several pests, (4) the application mistakes during pesticide spraying (Waichman et al., 2007; Darko & Akoto, 2008). Recently, European markets are requesting particular specifications such as application of pesticide residues below MRL as well as limitations for multi-residues and indexes for acute and chronic risk assessments. Although the establishment of MRLs is based on good agricultural practices (GAP) data on fresh foods derived from commodities, these are not toxicological limits (Blasco et al., 2006). Nevertheless, exceedance of MRLs is significant violations of GAP, and MRLs can not be considered as reliable tools for the assessment of the acute and chronic risks alone. Therefore, dietary risk assessment of insecticides has recently gained a great attention (Nasreddine & Parent-Massin, 2002; Gebara et al., 2011; Marete et al., 2020; Chen et al., 2011; Balkan & Yılmaz, 2022b). The long term (chronic) dietary risk assessments are made based on daily food consumption and detected pesticide residue data on each commodity. Then, the estimated chronic dietary exposure is compared with the acceptable daily intake (ADI) value which gives the concentration of a chemical that can be consumed over a long period without adverse health effects. For the short-term (acute) dietary risk assessment, the Acute Reference Dose (ARfD) is used to identify possible consumer health risks. The ARfD gives the concentration of a chemical that can be ingested over a short period of time (one meal, one day) without significant risks. For acute assessments one should focus on the edible portion of food commodities on the market, whereas for chronic assessments one should focus on raw agricultural commodities (Brancato et al., 2018).

Some commodities, namely carrot, lettuce, parsley, dill, apple, banana, pear, mandarin and orange are commonly consumed as main fruits and vegetables for Turkish consumers during the winter season. Therefore, assessing the risk of pesticide residues in these commodities intended for human consumption is necessary. One of the significant parameters in the evaluation of acute or chronic dietary risks is the frequency of exposure. The more the consumer is exposed to the chemical, the faced risk is higher. For this reason, in this study, it is desired to focus on the fruits and vegetables that people living in Bursa province consume frequently during the winter period. For this purpose, 223 people were asked about their consumption preferences in the winter period before the study. According to the results of the survey, the most commonly consumed items among Bursa consumers are 5 fruits (apple, pear, banana, mandarin and orange) and 4 vegetables (lettuce, parsley, dill and carrot), which were accepted as the research material. This study,

which analyzes the exposure of consumers during the winter period, has a unique value in this respect. The aims of the current study were to investigate pesticide residues in widely consumed seasonal fruit and vegetable samples collected from the Bursa markets and to conduct acute and chronic health risk assessments for human, based on exposure to the detected residue concentrations determined in 5 fruits and 4 vegetable commodities.

# **Materials and Methods**

## Chemicals and reagents

Insecticide standarts (Dr. Ehrenstorfer GmbH, Wesel, Germany) and other solvents and reagents used are of analytical grade. Chemical and toxicological properties of acaricides and insecticides are shown in Table S1 (PPDB, 2023; EU Pesticide Database, 2023). Quick Easy Cheap Effective Rugged Safe (QuEChERS) extraction kits [6 g anhydrous magnesium sulfate (MgSO<sub>4</sub>) + 1 g anhydrous sodium acetate (NaOAC)] and clean-up kits [1.2 g MgSO<sub>4</sub>, 0.4 g primary and secondary amines (PSA, 40  $\mu$ m particle size) + 0.4 g C<sub>18</sub>] were used.

#### Instruments and LC-MS/MS conditions

LC-MS/MS device was used for chromatographic analyses (Agilent 1260 Infinity II HPLC System and Agilent 6470 Triple Quadrupole Liquid-Mass Spectrometry). The device is connected with Agilent Poroshell SB-C<sub>18</sub> (3 mm x 100 mm x 2.7 µm) column. Flow rate, injection volume and total run time were 0.5 mL/min, 1 µL and 15 minutes, respectively. Two mobile phases were used namely A (0.1% formic acid+1mM ammonium fomat in water and B (Metanol). Following gradient program is used: 0-0.05 min. 70% A; 8 min. 5%; 8-12.5 min. 5% A; 12.6 min. 70% A; 12.6-15 min 70% A. Retention times (tR), precursor ion and fragment ions of each acaricides and insecticides are given in Table 1. The other instruments used in the current study are blender (Retsch, GM 300), precise balance (Ohaous, AV812), centrifuge (OHAOUS, FC5706), orbital shaker (Biosan, PSU-10İ), vortex (FAITHFUL, MX-S), micropipets (Eppendorf, K49321I, L17301I, M32978I), and ultra pure water machine (MX-S).

#### Verification of the analysis

Verification studies were performed in an accredidated analysis laboratory based on the criteria of Analytical Quality Control and Method Validation Procedures for Pesticide Residues Analysis in Food and Feed SANTE 11312/2021, such as linearity, recovery, precision and limit of quantification (LOQ). Calibration (matrix match standards) was performed on blank tomato representing fresh vegetables and fruits (CAC, 2003; SANTE, 2021). Blank tomato samples of 1 kg were homogenized with a blender. For recovery tests, 15 g blank samples were spiked with 100 µL of insecticide spike solutions (in MeCN). Tests were conducted in five replicates (five replicate analytical portions). Linearity was evaluated using six levels ranging from 5 µg to 250 µg L<sup>-1</sup> prepared with MeCN. Matrix matched calibration curve was used to quantify insecticides. Recovery and precision parameters were determined for two spiking concentrations (10 and 50 µg kg<sup>-1</sup>) across five different time points and by two different analysts. Calibration analysis results, retention times (tR) and selected ion groups of the analyzed insecticides were given in Table 1. Matrixmatched calibration curves of the 38 insecticides were linear (R<sup>2</sup> = 0.998-0.999). The retention times (tR) ranged between 0.99-10.83 min. The regression equations of the matrix-matched calibration curves were used for quantification of the insecticides. Trueness and precision were assessed based on recovery, repeatability and reproducibility parameters (Tiryaki, 2016; SANTE, 2021). Detection limits (LODs), LOQs, recovery rates (%) and relative standart deviations for repeatability and reproducibility (RSD<sub>r</sub> and RSD<sub>wr</sub> %) of all insecticides were found compatible with SANTE 2021 criteria. The LOQ values were quite lower than the MRLs of each insectides (Table 5). The recovery rates of the insecticides for two spike levels were calculated between 90.46-117.41 and 96.16-115.55, respectively. The highest RSD<sub>r</sub> and RSD<sub>wr</sub> were 12.64 and 17.56 for 10 µg kg<sup>-1</sup> and 7.50 and 8.30 for 50 µg kg<sup>-1</sup> respectively. All vertification parameters were compatible with SANTE 11312/2021 criteria (SANTE, 2021).

Table 1. Calibration analysis results, retention times (tR) and selected ion groups and their collision energies of the analyzed pesticides

Pesticide	tR* min	Calibration equation y=a+bx	Determination co-efficient, R <sup>2</sup>	Precursor ion, m/z (CE**)	Fragment ion, m/z (CE)
Acetamiprid	2.67	y=18094.5x+15269.9	0.9999	223.1	126.1 (17), 56.2 (11)
Abamectin	9.95	y=466.678172x+480.11	0.9993	895.2	327.3 (50), 449.3 (48)
Bifenazate	7.45	y=21034.5x+9728.6	0.9996	301.2	170.0 (20), 198.0 (5)
Bifenthrin	10.25	y=2251.4x+2674.5	0.9992	440.2	166.1 (20), 181.1 (7)
Chlorantraniliprole	5.73	y=1256.6x+1590.5	0.9994	484.0	283.9 (21), 285.9 (21)
Chlorfenvinphos	8.21	y=853.35x+6375.48	0.9996	359.1	155.1 (7), 99.1 (29)
Chlorpyrifos	9.26	y=4163.3x+827.61	0.9997	351.9	199.9 (15), 197.9 (15)
Chlorpyrifos methyl	8.58	y=1450.84x-511.92	0.9980	321.9	125 (17), 289.9 (11)
Clofentezine	8.39	y=7331.31x-3939.88	0.9988	303.1	102.1 (37), 138.1 (9)
Clothianidin	2.71	y=1586.6x-1078.8	0.9993	250.1	132 (15), 169.1 (13)
Cypermethrin	8.77	y=805.29x-670.28	0.9994	433.0	126.8 (34), 191.0 (12)
Cyromazine	0.99	y=12507.8x-1505.7	0.9994	167.3	85.2 (17), 125.2 (15)
Deltamethrin	8.76	y=436.67x-846.63	0.9996	522.8	280.6 (12), 505.8 (6)
Diflubenzuron	7.81	y=1993.8x+2330.67	0.9997	310.9	141 (15), 158 (6)
Emamectin B1a	8.81	y=18167.36x-2108.51	0.9991	886.5	126.0 (40), 158.0 (40)
Ethoprophos	7.68	y=13696.7x+12063.0	0.9998	243.0	130.9 (20), 172.9 (10)
Etoxazole	9.37	y=12903.5x+8505.09	0.9996	360.0	113.0 (23), 141.0 (15)
Fenbutatin oxide	10.83	y= -3261.8x-4249.4	0.9987	519.3	197 (55), 351.1 (35)
Fenvalerate	9.67	y=252.2x+37.99	0.9996	439.0	167 (14), 169 (10)
Flubendiamide	7.91	y=2762.65x+3704.33	0.9994	681.0	253.9 (40), 273.9 (24)
Imidacloprid	2.54	y=2626.98x+2617.6	0.9994	256.1	175.0 (12), 209.0 (10)
Indoxacarb	8.55	y=708.68x+67.82	0.9990	528.1	150.0 (16), 203.0 (36)
Lambda cyhalothrin	7.88	y=425.78x-65.61	0.9993	467.1	225.0 (14), 450.0 (6)
Malathion	6.33	y=6662.9x-116.79	0.9995	330.9	127.0 (4), 285.0 (38)
Metaflumizone	8.90	y=9023.9x+25059.9	0.9998	505.0	117.0 (48), 302.0 (10)
Methoxyfenozide	7.23	y=8824.6x-2323.0	0.9993	369.1	133.1 (28), 149 (14)
Novaluron	8.65	y=1028.11x-1218.79	0.9975	492.7	140.7 (46), 158.0 (12)
Phosmet	6.64	y=950.85x+292.58	0.9998	317.9	133 (28), 160 (21)
Primicarb	4.88	y=21360.3x-20650.8	0.9995	239.2	72.1 (15), 182.1 (11)
Pirimiphos methyl	8.40	y=45364.5x-2259.3	0.9981	360.2	108.1 (31), 164.1 (19)
Pyridaben	9.75	y=34981.4x+19361.8	0.9988	365.2	147.1 (23), 309.1 (7)
Pyriproxyfen	9.18	y=50164.4x+21561.5	0.9992	322.2	96.1 (11), 185.0 (19)
Spinosad	7.32	y=3266.3x-3304.6	0.9989	732.5	98.2 (55), 142.1 (35)
Spirodiclofen	8.66	y= 5572x-8635.00	0.9985	411.0	71.0 (16), 313.0 (11)
Spirotetramat	7.54	y= 4124.6x-1872.5	0.9992	374.1	302.1 (23), 330.1 (21)
Quinalphos	8.49	y= 5142.1x-3133.1	0.9997	146.1	91 (24), 118 (10)
Tau fluvalinate	8.92	y=2080.2x+3139.7	0.9997	503.1	181.1 (25), 208.1 (15)
Thiacloprid	3.09	y=18017.3x-13317.1	0.9994	253.0	90.0 (35), 126.0 (16)

<sup>\*</sup>tR, retention time (min); \*\* CE, Collision Energy (V)

#### Consumer surveys

Both online and face to face questionnaire surveys applied in Bursa province between November 2022 and February 2023. For online surveys, the google form link was shared via mails and various social media networks. The survey consisted of 223 respondents. The respondents consisted of 68% females and 32% males (age 16 to 70) and the largest proportion (74%) was comprised of middle-aged respondents (age 23 to 45). Mean body weight of female and male respondents determined as 64.86 kg and 81.16 kg respectively (female and male mean body weight 70.05 kg). The survey questions were provided in the Table 2.

Table 2. Questions in consumer questionnaire surveys

Questions		Ans	wers
Do you consume X commodity?		Yes:	No:
If yes; What is your consumption frequency?	everyday day:	per week:	days per month:
Specify your individual daily consumption amount for X comm	odity in portions:	;	oortions*
Specify the maximum amount of X commodity that you can cor	nsume at one time		portions

<sup>\*</sup> The following data were used in the grammatical translation of the survey results (TÜBER 2019):

#### Collecting samples

The agricultural commodity samples, namely carrot, lettuce, parsley, dill, apple, banana, pear, mandarin and orange, were collected from different local open markets and supermarkets of Bursa province for 4 weeks during February 2023. Each commodity sample (totally 99) of about 1 kg were homogenized and 15 g analytical portions (in triplicates) were obtained for the analysis. Extraction and cleaning procedures are shown in Figure 1 (Lehotay, 2007). Spiked and collected samples were analysed in LC-MS/MS system.

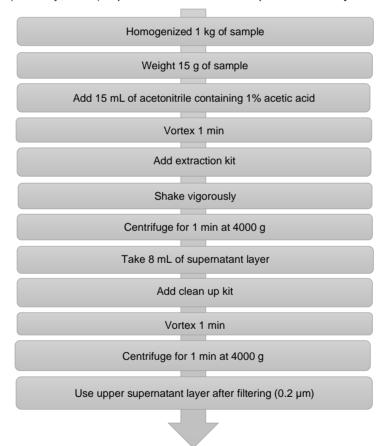


Figure 1. Analytical steps for extraction and cleaning (QuEChERS-AOAC Official Method 2007.01).

# Methodology for assessing dietary intake of insecticides

Estimation of acute and chronic risks to consumer health were performed based on the previous studies (Chen et al., 2011; Kazar Soydan et al., 2021). The dietary exposure to insecticides has been calculated in order to assess the acute consumer health risk for adults.

<sup>2</sup> cups or 2 fists or 1 large bowl = 1 standard portion=75 g of dill; 2 cups or 2 punches or 1 large bowl = 1 standard portion= 75 g of lettuce/or parsley; 1 medium size or 1 cup or 1 punch = 1 standard portion= 150gr of carrots; 1 medium size; 7 cm in diameter or 1 fist size = 1 standard portion= 150gr apples/oranges; 1 small size or 5 pieces = 1 standard portion= 150gr pears; 2 medium size-6 cm diameter = 1 standard portion= 150gr mandarins; 1 hand length or sliced 2/3 small bowl= 100g of banana.

The following input values are required to calculate the actual acute exposure:

- Maximum residue level of each insecticide obtained from analysis of the above-mentioned samples of 5 fruits and 4 vegetables in 2023
- Annual fruit/vegetable consumptions per person (97.5th percentile of eaters) were determined based on the survey results of present study (Table 3).
- The average body weight of an adult is taken as 70 kg based on our survey and TUIK data (2022).

The estimated short-term intake (ESTI) was calculated based on the following formula:

$$ESTI = \frac{LP \ x \ MRL \ x \ CF \ x \ PF \ x \ VF}{BW} \tag{1}$$

Where, *LP*, Large portion reported (kg day<sup>-1</sup>) (97.5th percentile of eaters); *MR*L, Maximum residue level for each commodity (mg kg<sup>-1</sup>); *CF*, Conversion factor residue definition enforcement to residue definition risk assessment; *PF*, Processing factor or peeling factor; *VF*, Variability factor was used as 7 for orange, mandarin, apple, pear, banana; 5 for carrot and lettuce; 1 for parsley and dill according to Brancato et al. (2018); *BW*, mean body weight for the subgroup of the population related to mean consumption (kg).

An estimate of pesticide intake in the diet was compared to the ARfD of each insecticide (Table S1). The acute hazard index (aHI) was calculated as follows:

$$aHI = \frac{ESTI}{ARfD} \tag{2}$$

aHI ≤ 100% indicates that adverse effects are not likely to occur and thus can be considered to have negligible hazard.

The dietary exposure to pesticides has been calculated in order to assess the chronic consumer health risk for the adults indicated in EFSA PRIMo revision 3 (Brancato et al., 2018).

The estimated daily intake (EDI) of pesticide residues was calculated with the following formulas:

$$Cp, f = \frac{Cavg. pos. p. f \times Npos. p. f}{Np, f}$$
(3)

$$EDI = Cp_{,f} \cdot MCf$$
 (4)

Where:  $C_{p,f}$ , the average content (mg kg<sup>-1</sup>) of pesticide p in commodity f;  $C_{avg,pos,p,f}$ , the average content (mg kg<sup>-1</sup>) of pesticide p in commodity f with detected residues;  $N_{pos,p,f}$ , the number of samples with detected residues;  $N_{p,f}$ , the number of commodities analyzed for the pesticide EDI: the estimated daily intake (mg kg<sup>-1</sup> bw day<sup>-1</sup>) for each combination of pesticide p and commodity f;  $MC_f$ , is the average consumption rate of that commodity (g<sup>-1</sup> bw day<sup>-1</sup>) from obtained present study survey results.

The chronic risk assessment of intakes compared to pesticide toxicological data was performed by calculating the Chronic Hazard Quotient (cHQ) by dividing EDI by the relevant acceptable daily intake (ADI):

$$cHQ = \frac{EDI}{ADI} 100\% \tag{5}$$

The level of concern for cHQ value was set as 1. Therefore,  $cHQ \le 1$  indicates that adverse effects are not likely to occur and thus can be considered to have negligible hazard.

### **Results and Discussion**

# **Consumer survey**

The survey results on vegetable and fruit consumption behaviour of Bursa community were given in Table 3. This survey was conducted due to the lack of food consumption data for Turkish citizen and the obtained data was used in the assesment of chronic and acute dietary risks. Two significant data were provided from this survey for chronic and acute dietary risk assessments, respectively: (1) Daily consumption data (gr/person/day) and (2) Maximum consumption amount in a single meal (g/person). When daily consumption rates for each commodity were compared with the Turkish Statistical Institute (TUIK) database (TUIK, 2021), the values for carrot, pear and apple were similar, where as lettuce, orange, mandarin and banana values were higher. When the results were compared with the EU community, consumption rates for lettuce, parsley, orange and banana were similar but carrot, apple and pear consumption of Bursa community was lower (Brancato et al., 2018). Maximum consumption in single meal values could not be compared with the TUIK database since there is no data regarding the Turkish community. However, maximum consumption results for all commodites were found lower compared with those consumed by EU communities (Table 3). The discrepancy of consumption data from EU commodity, could be due to the differences in factors like consumption habits, geographical origin and availability of the product, nutrition regimes, enjoyment of food (Kapoor & Kar, 2022), and also sociodemographic characteristics such as age, gender, education and income (Mata et al., 2023). Since there is limited data about the comsumption habits of the Turkish community, a simple questionnaire like the one used in this research has upgraded the accuracy and reliability of the acute and chronic risk assessment for the community.

Table 3. Consumer preferences for fruits and vegetables in Bursa province

		Daily consumption (g/person/day)		Daily consumption (g/bw/day)*	in sin	consumption agle meal person)	Maximum consumption in single meal** (kg/day)
Commodity	Bursa community	EU community (Brancato et al., 2018)	Turkish community (TUIK, 2021)	Bursa community	Bursa community	EU community (Brancato et al., 2018)	Bursa community
Lettuce	25.03	36.69	14.79	0.357	93.35	159.80	0.09
Parsley	15.48	2.54	unknown	0.221	57.82	79.90	0.03
Dill	5.76	37.13	unknown	0.082	33.96	unknown	0.24
Carrot	52.93	29.93	14.25	0.756	238.61	259.40	0.06
Apple	61.60	202.18	85.45	0.879	217.22	664.00	0.22
Pear	13.65	44.26	13.15	0.195	121.66	781.70	0.12
Orange	67.57	65.124	26.85	0.965	263.68	996.50	0.26
Mandarin	75.28	10.64	21.09	1.075	283.30	720.94	0.28
Banana	48.83	54.78	24.93	0.697	184.53	611.00	0.18

\*MCf, is the average consumption rate of that commodity; \*\*LP, Large portion.

## Residues in the different commodities

The co-occurrence of insecticide residues is given in detail in Table 4. Among fruit samples, highest rate of samples with insecticide residues were calculated in apple (100%), pear (90.91%) and lettuce (90.91%). Except banana samples, residues of two or more insecticides were found in all other commodities. The five commodities, namely parsley, dill, apple, pear and mandarin, contained 4 and more insecticide residues with the ratios of 18.18, 36.36, 36.36, 36.36 and 27.27%, respectively. Three commodities, such as, dill, pear and mandarin, were contaminated with seven pesticide residues (with 9.1, 18.2 and 9.1%, respectively). Similarly, survey studies conducted in other countries reported presence of multiple pesticide residues (four or more) in different commodities such as pear, parsley, mandarin, orange, banana, apple (Chen, 2011; Ersoy et al., 2011; Esturk et al., 2014; Al-Shamary et al., 2016, El Hawari et al., 2019; Al-Nasir et al., 2020; Kazar Soydan et al., 2021; Kottadiyil et al., 2023). In accordance with previous studies,

the most frequent combinations of pesticides detected in the same sample were acetamiprid, cypermethrin, deltamethrin and imidacloprid (Chen, 2011; Ersoy et al., 2011; Jallow et al., 2017; El Hawari et al., 2019; Kumari, 2019; Kazar Soydan et al., 2021; Kottadiyil et al., 2023).

Table 4. Number of samples with multiple insecticide residues for each commodity

Commodity			Rate	of sample	es with mu	Itiple resid	ues (%)		
	0	1	2	3	4	5	6	7	Total (%)
Lettuce	9.09	63.64	27.27	-	-	-	-	-	90.99
Parsley	54.55	9.09	-	18.18	9.09	9.09	-	-	45.45
Dill	36.36	9.09	9.09	9.09	18.18	9.09	-	9.09	63.64
Carrot	63.64	27.27	9.09	-	-	-	-	-	36.36
Apple	-	-	36.36	27.27	18.18	9.09	9.09		100.00
Pear	9.09	18.18	9.09	27.27	9.09	9.09	-	18.18	90.99
Orange	72.73	9.09	18.18	-	-	-	-	-	27.27
Mandarin	45.45	18.18	-	9.09	18.18	-	-	9.09	54.55
Banana	54.55	45.45	-	-	-	-	-	-	45.45

MRL levels of 38 insecticides for each commodity and MRL exceedance rate (fold) were given in Table 5. The most of the MRL levels were provided from Turkish Food Codex (TGK, 2021). Since some insecticides used in certain commodities in Türkiye are not registrated, their MRL levels were obtained from the EU authorities (EU Pesticide Database, 2023). In the present study, insecticide residues in some of samples exceeded their MRL levels. In our study, 16.2% of the samples exceeded the approved MRL levels of detected insecticide and acaricides. Considering the highest residue concentrations detected in the current study, fenbutatin oxide and imidacloprid residues in lettuce exceeded their MRLs 2.6 and 2.8 folds, respectively. Imidacloprid MRL exceedance was also reported in nectarin samples (Serbes & Tiryaki, 2023). In parsely, chlorpyrifos and pirimiphos methyl residues was detected above 1.30 and 5.75 folds of their MRLs, respectively. The highest MRL exceedance was observed in dill with cypermethrin (1.26 folds), ethoprophos (4.85 folds), imidacloprid (1.26 folds), malathion (5.85 folds) and spirotetramat (1.08 folds). In carrot, one of the two insecticides exceeded MRL level (Imidacloprid 2.80 folds). In fruits, there were relatively fewer instances of insecticides exceeding their MRL levels: diflubenzuron (4.50 folds) in pear; chlorpyrifos (9.50 folds) and fenvalerate (1.35 folds) in mandarin and tau-fluvalinate (2.90 folds) in banana (Table 6). Previous studies reported that 8.4-22% of fruit and vegetable samples contained pesticide residues above the approved MRL levels (Chen et al., 2011, EL-Saeid & Selim, 2013; Jallow et al., 2017; Mebdoua et al., 2017; Algharibeh & Al Fararjeh, 2019; Gondo et al., 2021; Balkan & Kara, 2022; Wang et al., 2022). Similarly, Estürk et al. (2014) and Balkan & Yılmaz (2022a) also reported MRL exceedance in some pesticides detected in lettuce, parsley and various leafy vegetables.

Table 5. MRLs of insecticides

Pesticide	LOQ	MRL (mg kg <sup>-1</sup> )*									
	(µg kg <sup>-1</sup> )	L	PA	D	С	Α	PE	0	M	В	
Acetamiprid	5.55	1.5	3.0	0.05	-	0.8	0.4	0.9	0.9	-	
Abamectin	4.37	-	-	0.05	-	-	0.03	-	-	-	
Bifenazate	6.50	-	-	-	-	0.7	-	-	-	-	
Bifenthrin	6.86	-	-	-	-	-	-	-	0.05	-	
Chlorantraniliprole	7.97	-	-	-	-	0.5	0.5	-	-	-	
Chlorfenvinphos	9.79	-	-	-	-	-	-	-	0.01	-	
Chlorpyrifos	6.97	-	0.01	-	-	-	-	-	0.01	-	
Chlorpyrifos methyl	9.03	-	-	0.01	0.04	-	-	-	-	-	
Clofentezine	8.03	-	-		-	0.5	-	-	-	-	
Clothianidin	6.57	-	-	0.2	-	-	-	-	-	-	
Cypermethrin	6.63	-	-	0.1	-	1	1.0	-	2.0	-	
Cyromazine	5.58	0.01	-		-	-	-	-	-	-	
Deltamethrin	7.39	0.5	2.0	0.1	-	0.2	0.1	-	-	-	
Diflubenzuron	9.68	-		-	-	5	0.01	-	-	-	
Emamectin B1a	6.63	-	0.2	-	-	-	-	-	-	-	
Ethoprophos	6.75	-	-	0.02	-	-	-	-	-	-	

Table 5. Continued

Pesticide	LOQ				MR	L (mg k	g <sup>-1</sup> )*			
Pesticide	(µg kg <sup>-1</sup> )	L	PA	D	С	Α	PE	0	М	В
Etoxazole	7.63	-	-	-	-	-	-	-	0.1	-
Fenbutatin oxide	7.08	0.01	0.02	-	-	-	-	-	-	-
Fenvalerate	9.04	-	-	-	-	0.05	-	-	0.02	-
Flubendiamide	6.93	-	-	-	-	8.0	-	-	-	-
Imidacloprid	5.68	0.01	0.05	0.05	0.01	-	0.5	-	-	-
Indoxacarb	9.95	-	-			0.5	-	-	-	-
Lambda cyhalothrin	7.90	-	-	0.3	-	-	0.08	0.2	-	-
Malathion	8.69	-	-	0.02	-	-	-	-	2.0	-
Metaflumizone	9.24	-	-	0.1	-	-	-	-	-	-
Methoxyfenozide	6.63	-	-	-	-	2	-	-	-	-
Novaluron	7.70	-	-	-	-	0.01	0.01	-	-	-
Phosmet	9.78	-	-	-	-		0.5	-	-	-
Pirimicarb	5.70	-	3.0	5.0		0.5	-	-	-	-
Pirimiphos methyl	5.96	-	0.02	3.0	-	-	-	-	-	-
Pyridaben	5.94	-	0.02			0.9	-	-	0.3	-
Pyriproxyfen	5.98	-	-	-	-	-	0.2	-	0.6	0.7
Spinosad	6.84	10	-	-	-	-	-	-		-
Spirodiclofen	6.20	-	-	-	-	8.0	0.8	-	0.4	-
Spirotetramat	7.91	-	4.0	0.1		-	-	1.0	-	-
Quinalphos	7.36	-	-	-	-	-	-	0.01	-	0.01
Tau fluvalinate	8.58	-	-	-	-	0.3		0.4	0.4	-
Thiacloprid	5.53	-	-	-	-	0.3	0.3	-	-	-

<sup>\*</sup>MRL levels were obtained from TGK or from EU database: -: not detected in this commodity, L: lettuce, PA: parsely, D: dill, C: carrot, A: apple, PE: pear, O: orange, M: mandarin, B: banana.

Table 6. MRL exceedance rate of the highest insecticide residues

Pesticide			M		edance	rate (fol	d)		
Pesticide	L	PA	D	С	Α	PE	0	М	В
Acetamiprid	0.11	0.03	5.44	-	0.02	0.04	0.01	0.01	-
Abamectin	-	-	0.32	-	-	0.17	-	-	-
Bifenazate	-	-	-	-	0.02	-	-	-	-
Bifenthrin	-	-	-	-	-	-	-	0.88	-
Chlorantraniliprole	-	-	-	-	0.04	0.02	-	-	-
Chlorfenvinphos	-	-	-	-	-	-	-	0.90	-
Chlorpyrifos	-	1.30	-	-	-	-	-	9.50	-
Chlorpyrifos methyl	-	-	4.10	0.53	-	-	-	-	-
Clofentezine	-	-		-	0.09	-	-	-	-
Clothianidin	-	-	0.04	-	-	-	-	-	-
Cypermethrin	-	-	1.26	-	0.02	0.01	-	0.01	-
Cyromazine	0.40	-	-	-	-	-	-	-	-
Deltamethrin	0.13	0.01	0.21	-	0.03	0.05	-	-	-
Diflubenzuron	-	-	-	-	0.01	4.50	-	-	-
Emamectin B1a	-	0.22	-	-	-	-	-	-	-
Ethoprophos	-	-	4.85	-	-	-	-	-	-
Etoxazole	-	-	-	-	-	-	-	0.05	-
Fenbutatin oxide	2.60	0.35	-	-	-	-	-	-	-
Fenvalerate	-	-	-	-	0.08	-	-	1.35	-
Flubendiamide	-	-	-	-	0.01	-	-	-	-
Imidacloprid	2.80	0.18	1.26	2.80	-	0.09	-	-	-
Indoxacarb	-	-	-	-	0.05	-	-	-	-
Lambda cyhalothrin	-	-	0.17	-	-	0.55	0.13	-	-
Malathion	-	-	5.85	-	-	-	-	0.08	-
Metaflumizone	-	-	0.26	-	-	-	-	-	-
Methoxyfenozide	-	-	-	-	0.01	-	-	-	-
Novaluron	-	-	-	-	1.00	0.70	-	-	-
Phosmet	-	-	-	-	-	0.03	-	-	-
Pirimicarb	-	0.01	0.08	-	0.02	-	-	-	-
Pirimiphos methyl	-	5.75	0.17	-	-	-	-	-	-
Pyridaben	-	0.75	-	-	0.02	-	-	0.06	-
Pyriproxyfen	-	-	-	-	-	0.33	-	0.01	0.01
Spinosad	0.05	-	-	-	-	-	-	-	-
Spirodiclofen	-	-	-	-	0.01	0.07	-	0.01	-
Spirotetramat	-	0.01	1.08	-	-	-	0.01	-	-
Quinalphos	-	-	-	-	-	-	0.40	-	2.90
Tau fluvalinate	-	-	-	-	0.01	-	0.02	0.03	-
Thiacloprid	-	-	-	-	0.09	0.15	-	-	

<sup>-,</sup> not detected in this commodity: L: Lettuce, PA: parsely, D: dill, C: carrot, A: apple, PE: pear, O: orange, M: mandarin, B: banana.

### Chronic and acute dietary risk assessments in different commodities

For the risk assessment of insecticide and acaricide residues in each commodity, Cpf (the average content of pesticide p in commodity f) and HR (highest residue) for fruits and vegetables were given in Tables 7 and 8 respectively. The ARfD and ADI values for each pesticide were previously given in Table S1. The other important parametres, daily consumption (MCf) and maximum consumption in single meal (LP) were also shown in Table 3. Using all these parameters, the estimated daily intake (EDI) for chronic risk and the estimated short-term intake (ESTI) for acute risk were calculated. Thus, the chronic hazard quotient (cHQ) and acute hazard index (aHI) for adults were listed in Tables S2 and S3. According to the findings of the current study, the chronic hazard was not observed for any of the insecticides in all commodities. The cHQ of many pesticides were close to zero or <0.010. The highest cHQ values were 0.1286 for emamectin B1a in parsley, 0.1813 for ethoprophos in dill and 0.1368 in chlorfenvinphos for mandarin. Moreover, when the cumulative long-term exposure (total cHQ) was evaluated, none of the insecticides was found risky for adults.

Table 7. Mean and highest insecticide residue levels detected in fruits

					Comm					
Pesticide	Ар	ple	Pe	ear		inge	Man			ana
	Cp.f	HR								
Acetamiprid	0.001	0.019	0.001	0.017	0.001	0.012	0.001	0.005	-	-
Abamectin	-	-	0.001	0.005	-	-	-	-	-	-
Bifenazate	0.001	0.011	-	-	-	-	-	-	-	-
Bifenthrin	-	-	-	-	-	-	0.004	0.044	-	-
Chlorantraniliprole	0.001	0.019	0.001	0.009	-	-	-	-	-	-
Chlorfenvinphos	-	-	-	-	-	-	0.001	0.009	-	-
Chlorpyrifos	-	-	-	-	-	-	0.009	0.095	-	-
Chlorpyrifos methyl	-	-	-	-	-	-	-	-	-	-
Clofentezine	0.004	0.044	-	-	-	-	-	-	-	-
Clothianidin	-	-	-	-	-	-	-	-	-	-
Cypermethrin	0.001	0.017	0.001	0.008	-	-	0.001	0.008	-	-
Cyromazine	-	-	-	-	-	-	-	-	-	-
Deltamethrin	0.001	0.006	0.001	0.005	-	-	-	-	-	-
Diflubenzuron	0.005	0.057	0.005	0.045		-	-	-	-	-
Emamectin B1a	-	-	-	-	-	-	-	-	-	-
Ethoprophos	-	-	-	-	-	-	-	-	-	-
Etoxazole	-	-	-		-	-	0.001	0.005	-	-
Fenbutatin oxide	-	-	-	-	-	-	-	-	-	-
Fenvalerate	0.001	0.004	-	-	-	-	0.003	0.027	-	-
Flubendiamide	0.001	0.011	-	-	-	-	-	-	-	-
Imidacloprid	-	-	0.003	0.047	-	-	-	-	-	-
Indoxacarb	0.001	0.024	-	-	-	-	-	-	-	-
Lambda cyhalothrin	-	-	0.002	0.044	0.002	0.026	-	-	-	-
Malathion	-	-	-	-	-	-	0.006	0.155	-	-
Metaflumizone	-	-	_	-	-	-	_	-	-	_
Methoxyfenozide	0.001	0.015	-	-	-	-	-	-	-	-
Novaluron	0.001	0.01	0.001	0.007	-	-	_	-	-	_
Phosmet	-	-	0.001	0.013	-	-	_	-	-	_
Pirimicarb	0.001	0.009	-	-	-	-	_	-	-	_
Pirimiphos methyl	-	-	_	-	-	-	_	-	-	_
Pyridaben	0.001	0.011	-	-	-	_	0.002	0.018	-	-
Pyriproxyfen	-	-	0.003	0.065	_	_	0.001	0.006	0.001	0.00
Spinosad	_	-	-	-	_	_	-	-	-	-
Spirodiclofen	0.001	0.008	0.003	0.052	_	_	0.001	0.005	_	_
Spirotetramat	-	-	-	-	0.001	0.010	-	-	_	_
Quinalphos	_	-	-	_	0.001	0.004	_	-	0.002	0.029
Tau fluvalinate	0.001	0.004	_	_	0.001	0.004	0.001	0.013	-	-
Thiacloprid	0.001	0.029	0.003	0.044	-	-	-	-	_	_

Table 8. Mean and highest insecticide residue levels detected in vegetables

				Comi	nodity			
Pesticide	Lett	uce	Par	sley	D	ill	Ca	rrot
	Cp.f	HR	Cp.f	HR	Cp.f	HR	Cp.f	HR
Acetamiprid	0.009	0.171	0.007	0.103	0.014	0.272	-	-
Abamectin	-	-	-	-	0.002	0.016	-	-
Bifenazate	-	-	-	-	-	-	-	-
Bifenthrin	-	-	-	-	-	-	-	-
Chlorantraniliprole	-	-	-	-	-	-	-	-
Chlorfenvinphos	-	-	-	-	-	-	-	-
Chlorpyrifos	-	-	0.001	0.013	-	-	-	-
Chlorpyrifos methyl	-	-	-	-	0.002	0.041	0.002	0.021
Clofentezine	-	-	-	-	-	-	-	-
Clothianidin	-	-	-	-	0.001	0.008	-	-
Cypermethrin	-	-	-	-	0.012	0.126	-	-
Cyromazine	0.001	0.004	-	-	-	-	-	-
Deltamethrin	0.002	0.066	0.002	0.024	0.001	0.021	-	-
Diflubenzuron	-	-	-	-	-	-	-	-
Emamectin B1a	-	-	0.003	0.043	-	-	-	-
Ethoprophos	-	-	-	-	0.009	0.097	-	-
Etoxazole	-	-	-	-	-	-	-	-
Fenbutatin oxide	0.0024	0.026	0.001	0.007	-	-	-	-
Fenvalerate	-	-	-		-	-	-	-
Flubendiamide	-	-	-		-	-	-	-
Imidacloprid	0.003	0.028	0.001	0.009	0.006	0.063	0.002	0.028
Indoxacarb	-	-	-		-	-	-	-
Lambda cyhalothrin	-	-	-	-	0.004	0.05	-	-
Malathion	-	-	-		0.006	0.117	-	-
Metaflumizone	-	-	-		0.002	0.026	-	-
Methoxyfenozide	-	-	-		-	-	-	-
Novaluron	-	-	-		-	-	-	-
Phosmet	-	-	-		-	-	-	-
Pirimicarb	-	-	0.003	0.029	0.021	0.396	-	-
Pirimiphos methyl	-	-	0.006	0.115	0.045	0.496	-	-
Pyridaben	-	-	0.001	0.015	-	-	-	-
Pyriproxyfen	-	-	-		-	-	-	-
Spinosad	0.0424	0.466	-		-	-	-	-
Spirodiclofen	-	-	-		-	-	-	-
Spirotetramat	-	-	0.002	0.025	0.009	0.108	-	-
Quinalphos	-	-	-		-	-	-	-
Tau fluvalinate	-	-	-		-	-	-	-
Thiacloprid	-	-	-		-	-	-	-

The highest total cHQ values were observed for chlorfenvinphos (0.1368), chlorpyrifos (0.1189), emamectin B1a (0.1286), ethoprophos (0.1813), lambda cyhalothrin (0.1233), pirimiphos methyl (0.1268) when all the commodities were considered together. Among these insecticides, chlorfenvinphos and chlorpyrifos were banned in Türkiye in 2010 and 2020, respectively (BKU, 2023). Similarly, the chronic risks for the detected residues of these insecticides were also found negligible for human health with the previous studies conducted with peach, apple, pepper, tomato and cucumber by different reserchers (Mebdoua et al., 2017; El Hawari et al., 2019; Camara et al., 2020; Catak & Tiryaki 2020; Dulger & Tiryaki 2021; Zhang et al., 2021). The highest acute hazard index values obtained with this study exceeded ARfD for adults and calculated as 104.27% for indoxacarb in apples, 158.2% for phosmet in pears and 107.06% and 137.11% for lambda cyhalothrin in pears and mandarin, respectively. Acute toxicity risks of indoxacarb, phosmet and lambda-cyhalothrin were also reported by different previous studies (Mebdoua et al., 2017; El Hawari et al., 2019). Based on the WHO hazard classification, indoxacarb, lambda-cyhalothrin and phosmet are moderately hazardous insecticides (Classs II, Table S1). Although the highest residues of some insecticides, namely chlorpyrifos, cypermethrin, diflubenzuron, ethoprophos, fenbutatin oxide, fenvalerate, imidacloprid,

tau-fluvalinate, malathion, pirimiphos methyl and spirotetramat exceeded their MRL levels, the risk assessment in the present study showed that there were no acute and chronic dietary risks for these agricultural commodities. Acute risk assessments of chlorantraniliprole, chlorfenvinphos, clofentezine, diflubenzuron, etoxazole, fenvalerate, novaluron, pyriproxyfen, spirodiclofen and quinalphos could not perform due to the lack of ARfD values of these compounds in the EU Pesticide and PPDB Databases (Table S1).

Although insecticide residues detected in some products in this study exceeded the MRL levels determined for them; none of these compounds displayed a serious health risk for the consumer. No chronic risk has been determined for any insecticide, either on a product basis or cumulatively. Acute dietary risks were calculated for only 3 crops and 3 insecticides. This has shown that risks may arise from time to time due to wrong agricultural practices in the field. For this reason, it is important for public health to carry out monitoring studies regularly and to reveal the risks, as in this study.

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#### References

- Al-Nasir, F. M., A. G. Jiries, G. J. Al-Rabadi, M. H. Alu'datt, C. C. Tranchant, S. A. Al-Dalain, N. Alrabadi, O.Y. Madanat & R. S. Al-Dmour, 2020. Determination of pesticide residues in selected citrus fruits and vegetables cultivated in the Jordan Valley. LWT, 123: 109005.
- Al-Shamary, N. M., M. A. Al-Ghouti, I. Al-Shaikh, S. H. Al-Meer & T. A. Ahmad, 2016. Evaluation of pesticide residues of organochlorine in vegetables and fruits in Qatar: statistical analysis. Environmental Monitoring and Assessment, 188 (3): 1-14.
- Algharibeh, G. R. & M. S. Al Fararjeh, 2019. Pesticide residues in fruits and vegetables in Jordan using liquid chromatography/tandem mass spectrometry. Food Additives & Contaminants: Part B, 12 (1): 65-73.
- Ambrus, Á., V. V. N. Doan, J. Szenczi-Cseh, H. Szemánné-Dobrik & A. Vásárhelyi 2023. Quality control of pesticide residue measurements and evaluation of their results. Molecules, 28 (3): 954.
- Baldi, I., L. Filleul, B. Mohammed-Brahim, C. Fabrigoule, J. F. Dartigues, S. Schwall, J. P. Drevet, R. Salamon & P. Brochard, 2001. Neuropsychologic effects of long-term exposure to pesticides: results from the French Phytoner study. Environmental Health Perspectives, 109 (8): 839-844.
- Balkan, T. & K. Kara, 2022. Determination of pesticide residues and risk assessment in some vegetables grown in Tokat province. Plant Protection Bulletin, 62 (2): 26-35.
- Balkan, T. & Ö. Yılmaz, 2022a. Method validation, residue and risk assessment of 260 pesticides in some leafy vegetables using liquid chromatography coupled to tandem mass spectrometry. Food Chemistry, 384: 132516.
- Balkan, T. & Ö. Yılmaz, 2022b. Investigation of insecticide residues in potato grown in Türkiye by LC-MS/MS and GC-MS and health risk assessment. Turkish Journal of Entomology, 46 (4): 481-500.
- Berrada, H., M. Fernández, M. J. Ruiz, J. C. Moltó, J. Mañes & G. Font, 2010. Surveillance of pesticide residues in fruits from Valencia during twenty months (2004/05). Food Control, 21 (1): 36-44.
- Bhanti, M. & A. Taneja, 2007. Contamination of vegetables of different seasons with organophosphorous pesticides and related health risk assessment in northern India. Chemosphere, 69 (1): 63-68.
- BKU, 2023. Plant Protection Products Database. Republic of Turkey, Ministry of Agriculture and Forest, General Directorate of Food and Control, Department of Plant Protection Products. (Web page: https://bku.tarim.gov.tr) (Date accessed: November 2023) (in Turkish).
- Blasco, C., G. Font & Y. Picó, 2006. Evaluation of 10 pesticide residues in oranges and tangerines from Valencia (Spain). Food Control, 17 (11): 841-846.

- Brancato, A., D. Brocca, L. Ferreira, L. Greco, S. Jarrah, R. Leuschner, P. Medina, I. Miron, A. Nougadere, R. Pedersen, H. Reich, M. Santos, A. Stanek, J. Tarazona, A. Theobald & L. Villamar-Bouza, 2018. Guidance on use of EFSA Pesticide Residue Intake Model (EFSA PRIMo revision 3). EFSA Journal, 16 (1): 5147-5160.
- CAC, 2003. Representative commodities/samples for validation of analytical procedures for pesticide residues. In codex alimentarius commission guidelines on good laboratory practice in pesticide residue analysis. CAC/GL 40-1993. (Web page: http://www.fao.org/input/download/standards/378/cxg\_040e.pdf) (Date accessed: January 2023)
- Calvert, G. M., W. T. Sanderson, M. Barnett, J. M. Blondell & L. N. Mehler, 2001. "Surveillance of Pesticide-related Illness and Injury in Humans, 603-641". In: Handbook of Pesticide Toxicology (Eds. R. Krieger, J. Doull, D. Ecobichon, E. Hodgson, L. Reiter & J. Ross), Academic Press New York, 2342 pp.
- Cámara, M. A., S. Cermeño, G. Martínez & J. Oliva, 2020. Removal residues of pesticides in apricot, peach and orange processed and dietary exposure assessment. Food Chemistry, 325 (30): 126936.
- Catak, H. & O. Tiryaki, 2020. Insecticide residue analyses in cucumbers sampled from Çanakkale open markets. Turkish Journal of Entomology, 44 (4): 449-460.
- Chen, C., Y. Qian, Q. Chen, C. Tao, C. Li & Y. Li, 2011. Evaluation of pesticide residues in fruits and vegetables from Xiamen, China. Food Control, 22 (7): 1114-1120.
- Darko, G. & O. Akoto, 2008. Dietary intake of organophosphorus pesticide residues through vegetables from Kumasi, Ghana. Food and Chemical Toxicology, 46 (12): 3703-3706.
- Dulger, H. & O. Tiryaki, 2021. Investigation of pesticide residues in peach and nectarine sampled from Çanakkale, Turkey, and consumer dietary risk assessment. Environmental Monitoring & Assessment, 193 (9): 561.
- El Hawari, K., S. Mokh, M. Al Iskandarani, W. Halloum & F. Jaber, 2019. Pesticide residues in Lebanese apples and health risk assessment. Food Additives & Contaminants: Part B, 12 (2): 81-89.
- El-Saeid, M. H. & M. T. Selim, 2013. Multiresidue analysis of 86 pesticides using gas chromatography mass spectrometry: II-nonleafy vegetables. Journal of Chemistry, 25 (2): 727149.
- Ersoy, N., Ö. Tatlı, S. Özcan, E. Evcil, L. Ş. Coşkun & E. Erdoğan, 2011. Bazı tropikal ve suptropikal meyve türlerinde pestisit kalıntıları. Selcuk Journal of Agriculture and Food Sciences, 25 (2): 81-88.
- Esturk, O., Y. Yakar & Z. Ayhan, 2014. Pesticide residue analysis in parsley, lettuce and spinach by LC-MS/MS. Journal of Food Science & Technology, 51 (3): 458-466.
- EU Pesticide Database, 2023. Pesticide Residues MRLs. Directorate General for Health & Consumers. (Web page: https://food.ec.europa.eu/plants/pesticides/eu-pesticides-database\_en) (Date accessed: January, 2023)
- FAO, 2021. FAOSTAT, Crop Data Base. Food and Agriculture Organization United Nations. (Web page: http://www.fao.org/faostat/en/#data/QC/visualize) (Date accessed: January, 2023).
- Ferretti, G., T. Bacchetti, A. Belleggia & D. Neri, 2010. Cherry antioxidants: from farm to table. Molecules, 15 (10): 6993-7005.
- Gebara, A. B., C. H. P. Ciscato, S. H. Monteiro & G. S. Souza, 2011. Pesticide residues in some commodities: Dietary risk for children. Bulletin of Environmental Contamination and Toxicology, 86 (5): 506-510.
- Gondo, T. F., M. Kamakama, B. Oatametse, T. Samu, J. Bogopa & B. M. Keikotlhaile, 2021. Pesticide residues in fruits and vegetables from the southern part of Botswana. Food Additives & Contaminants: Part B, 14 (4): 271-280.
- Jallow, M. F. A., D. G. Awadh, M. S. Albaho, V. Y. Devi & N. Ahmad, 2017. Monitoring of pesticide residues in commonly used fruits and vegetables in Kuwait. International Journal of Environmental Research and Public Health, 14 (8): 833-845.
- Kapoor, P. & S. Kar, 2022. A Critical Evaluation of the Consumer Confidence Survey from India. Economic Studies (Ikonomicheski Izsledvania), 31 (7): 172-198.
- Kazar Soydan, D., N. Turgut, M. Yalçın, C. Turgut & P. B. K. Karakus, 2021. Evaluation of pesticide residues in fruits and vegetables from the Aegean region of Turkey and assessment of risk to consumers. Environmental Science and Pollution Research, 28 (22): 27511-27519.
- Kottadiyil, D., T. Mehta, R. Thasale & P. Sivaperumal, 2023. Determination and dietary risk assessment of 52 pesticide residues in vegetable and fruit samples by GC-MS/MS and UHPLC-QTOF/MS from Gujarat, India. Journal of Food Composition and Analysis, 115: 104957.

- Kumari, D. & S. John, 2019. Health risk assessment of pesticide residues in fruits and vegetables from farms and markets of Western Indian Himalayan region. Chemosphere, 224 (1): 162-167.
- Lehotay, S. J., 2007. Pesticide residues in foods by acetonitrile extraction and partitioning with magnesium sulfate gas chro-matography/mass spectrometry and liquid chromatography/tandem mass spectrometry. Journal of AOAC International, 90 (2): 485-520.
- Lozowicka, B., 2015. Health risk for children and adults consuming apples with pesticide residue. Science of the Total Environment, 502 (1): 184-198.
- Marete, G. M., V. O. Shikuku, J. O. Lalah & V. W. Wekasa, 2020. Occurrence of pesticides residues in French beans, tomatoes, and kale in Kenya, and their human health risk indicators. Environmental Monitoring & Assessment, 192 (11): 692.
- Mata, J., P. Kadel, R. Frank & B. Schüz, 2023. Education-and income-related differences in processed meat consumption across Europe: The role of food-related attitudes. Appetite, 182: 106417.
- Mebdoua, S., M. Lazali, S. M. Ounane, S. Tellah, F. Nabi & G. Ounane, 2017. Evaluation of pesticide residues in fruits and vegetables from Algeria Part B Surveillance. Food Additives & Contaminants: Part B, 10 (2): 91-98.
- Nasreddine, L. & D. Parent-Massin, 2002. Food contamination by metals and pesticides in the European Union. Should we worry? Toxicology Letters, 127 (1-3): 29-41.
- PPDB, 2023. IUPAC Pesticides Properties DataBase. (Web page: http://sitem.herts.ac.uk/aeru/iupac/) (Date accessed: January 2023).
- Prior, R. L., 2003. Fruits and vegetables in the prevention of cellular oxidative damage. The American Journal of Clinical Nutrition, 78 (3): 570-578.
- SANTE, 2021. Analytical quality control and method validation procedures for pesticide residues analysis in food and feed: SANTE 11312/2021. (Web page: https://www.accredia.it/en/documento/guidance-sante-11312-2021-analytical-quality-control-and-method-validation-procedures-for-pesticide-residues-analysis-in-food-and-feed/) (Date accessed: January, 2023)
- Serbes, E. B. & O. Tiryaki, 2023. Determination of insecticide residues in "Bayramiç Beyazı" nectarines and their risk analysis for consumers. Turkish Journal of Entomology, 47 (1): 73-85.
- Solecki, R., L. Davies, V. Dellarco, I. Dewhurst, M. V. Raaij & A. Tritscher, 2005. Guidance on setting of acute reference dose (ARfD) for pesticides. Food and Chemical Toxicology, 43 (11): 1569-1593.
- TGK, 2021. Türk Gida Kodeksi Pestisitlerin Maksimum Kalinti Limitleri Yönetmeliği. Sayı: 31611, Ankara. (Web page: https://www.resmigazete.gov.tr/eskiler/2021/09/20210927M1-1.htm) (Data accessed: October 2023) (in Turkish).
- Tiryaki, O., 2016. Validation of QuEChERS method for the determination of some pesticide residues in two apple varieties. Journal Environmental Science and Health, Part B, 51 (10): 722-729.
- TÜBER, 2019. Türkiye Beslenme Rehberi T.C. Sağlık Bakanlığı Yayın No: 1031, Ankara. (Web page: https://hsgm.saglik.gov.tr/depo/birimler/saglikli-beslenme-hareketli-hayat-db/Türkiye\_Beslenme\_Rehberi\_TUBER\_18\_04\_2019.pdf) (Date accessed: January 2023) (in Turkish).
- TUİK, 2021. Turkish Statistical Institute. (Web page: https://biruni.tuik.gov.tr/medas/?kn=92&locale=tr) (Date accessed: January 2023) (in Turkish).
- Waichman, A. V., E. Eve & N. C. da Silva Nina, 2007. Do farmers understand the information displayed on pesticide product labels? A key question to reduce pesticides exposure and risk of poisoning in the Brazilian Amazon. Crop Protection, 26 (4): 576-583.
- Wang, R., Y. Yang, Y. Deng, D. Hu & P. Lu, 2022. Multiresidue analysis and dietary risk assessment of pesticides in eight minor vegetables from Guizhou, China. Food Chemistry, 380: 131863.
- Yousefi, S. H, Aslani, M. Shakerkhatibi, Y. Mohammadia & G. H. Safari, 2022. Combined health risk assessment of organophosphates pesticide residues in greenhouse cucumber in the Northwestern of Iran based on Monte Carlo Simulations. International Journal of Environmental Analytical Chemistry, 102 (1): 1-16.
- Zhang, Y., W. Si, L. Chen, G. Shen, B. Bai & C. Zhou, 2021. Determination and dietary risk assessment of 284 pesticide residues in local fruit cultivars in Shanghai, China. Scientific Reports, 11 (1): 9681.

# **Supplementary Tables**

Table S1. Chemical and toxicological properties of acaricides and insecticides

Pesticide	Mode of action	Acceptable daily intake (mgkg <sup>-1</sup> bwday <sup>-1</sup> )	Acute reference dose (mgkg <sup>-1</sup> bwday <sup>-1</sup> )	for mammals oral acute LD <sub>50</sub> (mg kg <sup>-1</sup> bwday <sup>-1</sup> )	for mammals dermal LD <sub>50</sub> (mgkg <sup>-1</sup> bwday <sup>-1</sup> )	for mammals inhalation LD <sub>50</sub> (mgkg <sup>-1</sup> bw)	WHO clasification
Acetamiprid	Insecticide	0.025	0.025	>1.15	2000	146	II
Abamectin	Acaricide/Insecticide	0.0012	0.005	8.7	1914	>0.021	Ш
Bifenazate	Acaricide	0.01	0.1	>4.4	2000	>5000	U
Bifenthrin	Acaricide	0.015	0.03	54.5	2000	1.01	Ш
Chlorantraniliprole	Insecticide	1.56	-	>5.1	5000	>5000	U
Chlorfenvinphos	Insecticide	0.0005	-	12	31	0.05	III
Chlorpyrifos	Insecticide	0.001	0.005	66	1250	0.1	III
Chlorpyrifosmethyl	Insecticide	0.01	0.1	>0.67	2000	5000	III
Clofentezine	Acaricide	0.02	-	>5200	2100	>5.2	III
Clothianidin	Insecticide	0.097	0.1	>500	2000	>5.54	III
Cypermethrin	Insecticide	0.05	0.2	3.56	2000	287	II
Cyromazine	Insecticide	0.06	0.1	3387	3100	>3.6	III
Deltamethrin	Insecticide	0.01	0.025	0.6	2000	87	Ш
Diflubenzuron	Insecticide	0.1	-	>4640	2000	>2.5	Ш
Emamectin B1a	Insecticide	0.01	0.01	0.582	439	81.5	NL
Ethoprophos	Insecticide	0.0004	0.01	40	7.9	0.123	Ш
Etoxazole	Acaricide	0.04	-	>1.09	2000	>5000	NL
Fenbutatin oxide	Acaricide	0.05	0.1	>3000	2000	0.046	Ш
Fenvalerate	Insecticide	0.02	-	451	1000	>0.101	Ш
Flubendiamide	Insecticide	0.017	0.1	>0.0069	2000	>2000	Ш
Imidacloprid	Insecticide	0.06	0.08	>0.069	5000	131	II
Indoxacarb	Insecticide	0.005	0.005	>4.2	5000	179	II
Lambdacyhalothrin	Insecticide	0.0025	0.005	0.066	632	56	II
Malathion	Insecticide	0.03	0.3	>5	2000	1778	Ш
Metaflumizone	Insecticide	0.03	0.13	>5.2	5000	>5000	NL
Methoxyfenozide	Insecticide	0.1	0.1	>5000	5000	>4.3	Ш
Novaluron	Insecticide	0.01	-	5.15	2000	>5000	U
Phosmet	Insecticide	0.01	0.045	113	1000	>1.52	II
Pirimicarb	Insecticide	0.035	0.1	142	2000	>0.75	Ш
Pirimiphos methyl	Insecticide	0.004	0.1	>4.7	2000	1414	II
Pyridaben	Acaricide	0.01	0.05	0.62	2000	161	II
Pyriproxyfen	Insecticide	0.1	1.0	>1.3	2000	>5000	U
Spinosad	Insecticide	0.024	0.1	>5.18	5000	>2000	III
Spirodiclofen	Acaricide	0.015	-	>5.03	2000	>2500	NL
Spirotetramat	Insecticide	0.05	1.0	>2000	2000	>4.18	III
Quinalphos	Insecticide	-	-	71	1750	0.45	III
Tau fluvalinate	Insecticide	0.005	0.05	>0.56	2000	546	III
Thiacloprid	Insecticide	0.01	0.03	>1.2	2000	177	II

Class II: Moderately hazardous; Class III: Slightly hazardous; NL: Not listed; U: Unlikely to present an acute hazard.

Table S2. Chronic risk assessments of insecticides for fruits and vegetables in Bursa province

	cHQ - long-term dietary risk (chronic)										
Pesticide	Lettuce	Parsley	Dill	Carrot	Apple	Pear	Orange	Mandarin	Banana	Total cHQ	
Acetamiprid	0.012	0.0059	0.0046	-	0.0037	0.0011	0.0042	0.0019	-	0.0334	
Abamectin	-	-	0.0470	-	-	0.0035	-	-	-	0.0505	
Bifenazate	-	-	-	-	0.0088	-	-	-	-	0.0088	
Bifenthrin	-	-	-	-	-	-	-	0.0286	-	0.0286	
Chlorantraniliprole	-	-	-	-	0.0001	0.0001	-	-	-	0.0002	
Chlorfenvinphos	-	-	-	-	-	-	-	0.1368	-	0.1368	
Chlorpyrifos	-	0.0261	-	-	-	-	-	0.0928	-	0.1189	
Chlorpyrifos methyl	-	-	0.0014	0.0140	-	-	-	-	-	0.0154	
Clofentezine	-	-	-	-	0.0176	-	-	-	-	0.0176	
Clothianidin	-	-	0.0001	-	-	-	-	-	-	0.0001	
Cypermethrin	-	-	0.0188	-	0.0216	0.0028	-	0.0156	-	0.0588	
Cyromazine	0.0002	-	-	-	-	-	-	-	-	0.0002	
Deltamethrin	0.0073	0.0033	0.0011	-	0.0048	0.0007	-	-	-	0.0172	
Diflubenzuron	-	-	-	-	0.0046	0.0009	-	-	-	0.0055	
Emamectin B1a	-	0.1286	-	-	-	-	-	-	-	0.1286	
Ethoprophos	-	-	0.1813	-	-	-	-	-	-	0.1813	
Etoxazole	-	-	-	-	-	-	-	0.0011	-	0.0011	
Fenbutatin oxide	0.0017	0.0003	-	-	-	-	-	-	-	0.0020	
Fenvalerate	-	-	-	-	0.0026	-	-	0.0211	-	0.0237	
Flubendiamide	-	-	-	-	0.0052	-	-	-	-	0.0052	
Imidacloprid	0.0015	0.0003	0.0008	0.0019	-	0.0009	-	-	-	0.0054	
Indoxacarb	-	-	-	-	0.0183	-	-	-	_	0.0183	
Lambda cyhalothrin	-	-	0.0144	-	-	0.0177	0.0912	-	_	0.1233	
Malathion	-	-	0.0015	-	-	-	-	0.0212	_	0.0227	
Metaflumizone	-	-	0.0019	-	-	-	-	-	-	0.0019	
Methoxyfenozide	-	-	-	-	0.0007	-	-	-	_	0.0007	
Novaluron	-	-	-	-	0.0079	0.0012	-	-	_	0.0091	
Phosmet	-	-	-	-	-	0.0230	-	-	_	0.0230	
Pirimicarb	-	0.0017	0.0049	-	0.0021	-	-	-	_	0.0087	
Pirimiphos methyl	-	0.0341	0.0927	-	-	-	-	-	_	0.1268	
Pyridaben	-	0.0030	-	-	0.0088	-	-	0.0176	_	0.0294	
Pyriproxyfen	-	-	-	-	-	0.0010	-	0.0025	0.0006	0.0035	
Spinosad	0.0631	-	-	-	-	-	_	-	-	0.0631	
Spirodiclofen	-	-	-	-	0.0037	0.0034	_	0.0033	-	0.0104	
Spirotetramat	-	0.0010	0.0017	-	-	-	0.0018	-	-	0.0045	
Quinalphos	_	-	-	_	_	_	*	_	*	-	
Tau fluvalinate	-	-	-	-	0.0064	-	0.0105	0.0244	-	0.0413	
Thiacloprid	-	-	-	-	0.0164	0.0049	-	-	-	0.0213	

ARfD and ADI values were taken from EU Pesticide Database (2023); -: Residue not detected in this commodity, \*: Not allocated for this insecticide, there was no specified ARfD and/or ADI in EU Pesticide Database (2023).

Table S3. Acute risk assessments of insecticides for fruits and vegetables in Bursa province

			aHI	-short-te	erm dieta	ry risk (a	cute)		
Pesticide	Lettuce	Parsley	ΠΩ	Carrot	Apple	Pear	Orange	Mandarin	Banana
Acetamiprid	45.61	3.40	5.28	-	16.51	8.27	12.66	5.67	-
Abamectin	-	-	1.55	-	-	12.17	-	-	-
Bifenazate	-	-	-	-	2.39	-	-	41.55	-
Bifenthrin	-	-	-	-	-	-	-	-	-
Chlorantraniliprole	-	-	-	-	*	*	-	-	-
Chlorfenvinphos	-	-	-	-	-	-	-	*	-
Chlorpyrifos	-	2.15	-	-	-	-	-	53.83	-
Chlorpyrifos methyl	-	-	0.19	5.01	-	-	-	-	-
Clofentezine	-	-	-	-	*	-	-	-	-
Clothianidin	-	-	0.04	-	-	-	-	-	-
Cypermethrin	-	-	12.23	-	73.86	19.47	-	45.33	-
Cyromazine	0.27	-	-	-	-	-	-	-	-
Deltamethrin	44.01	1.98	1.02	-	-	6.08	-	-	-
Diflubenzuron	-	-	-	-	*	*	-	-	-
Emamectin B1a	-	3.55	-	-	-	-	-	-	-
Ethoprophos	-	-	4.71	-	-	-	-	-	-
Etoxazole	-	-	-	-	-	-	-	*	-
Fenbutatin oxide	1.73	0.06	-	-	-	-	-	-	-
Fenvalerate	-	-	-	-	*	-	-	*	-
Flubendiamide	-	-	-	-	2.39	-	-	-	-
Imidacloprid	2.33	0.09	0.38	8.35	-	7.15	-	-	-
Indoxacarb	-	-	-	-	104.27	-	-	-	-
Lambda cyhalothrin	-	-	4.85	-	-	107.06	137.11	-	-
Malathion	-	-	0.19	-	-	-	-	14.64	-
Metaflumizone	-	-	0.09	-	-	-	-	-	-
Methoxyfenozide	-	_	_	-	3.26	-	_	-	-
Novaluron	-	-	-	-	*	*	-	-	-
Phosmet	-	_	_	-	-	158.2	_	-	-
Pirimicarb	-	0.24	1.92	-	1.95	-	-	-	-
Pirimiphos methyl	-	0.63	1.60	-	-	-	-	-	-
Pyridaben	-	0.25	-	-	4.78	-	-	10.19	-
Pyriproxyfen	-	-	-	-	-	0.79	_	0.16	0.09
Spinosad	31.07	-	-	-	-	-	-	-	-
Spirodiclofen	-	-	-	-	*	*	-	-	-
Spirotetramat	-	0.02	0.52	-	-	-	0.26	-	-
Quinalphos	-	-	-	-	-	-	*	-	*
Tau fluvalinate	-	-	-	-	1.74	-	3.16	7.37	-
Thiacloprid	-	-	-	-	31.49	26.77	-	-	-

ARfD and ADI values were taken from EU Pesticide Database (2023); -: Residue not detected in this commodity, \*: Not allocated for this insecticide, there was no specified ARfD and/or ADI in EU Pesticide Database (2023).