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ORIGINAL ARTICLE

Health risk assessment of pesticide residues in vegetables collected from Dakahlia, Egypt

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Abstract

It has already been well established that long exposure to low doses of pesticides is linked to consumer risks. So, this study purposed to investigate the amounts of pesticide residues and potential health risks associated with them. The risk assessment was determined by two methods: 1. Pesticide toxicity index (PTI) depending on the maximum residue limit (MRL) to calculate the hazard quotient (HQ); 2. Health risk assessment (HR) using acceptable daily intake (ADI) and estimated daily intake (EDI) to calculate the health index (HI). Pesticide residues were estimated in 176 samples of the most popularly consumed vegetables collected from major retailers and markets in Dakahlia, Egypt (during 2018). There were 111 samples contaminated with pesticide residues (63.1%), of which 29 samples (16.48%) were higher than the maximum residue limits (MRL). Residues of 23 compounds were found in the analyzed samples, of which chlorpyrifos was the most frequent in 33 samples (18.75%); while cypermethrin was the lowest (detected in one sample). According to WHO toxicity classification, 12 of the detected pesticides were moderately hazardous (class II), seven pesticides belonged to class III (slightly hazardous), three compounds were found in class U (unlikely to pose an acute hazard with normal use), while carbofuran is a highly toxic compound (class Ib). Also, the obtained data revealed that, the HI's for the individual pesticides ranged from 0.0018 to 64.0% of ADI indicates no risk of adverse effects following exposure to the individual pesticides. The cumulative exposure amounts (PTI values) ranged from 1.58 in snake cucumber to 128.44 in potato tubers, indicating that, the combined risk index of pesticide residues was a significant health risk for consumers according to the individual risk index. It can be concluded that there is a need for strict regulation and regular monitoring of pesticide residues in foodstuff for consumers' health protection.

Keywords: pesticides residues, risk assessment, vegetables

Introduction

Nutrients and minerals present in vegetables and fruits are important for a healthy diet and play an important role in the prevention of chronic diseases. According to the World Health Organization, an average of 30% (based on mass) of food consumption consists of fruits and vegetables (WHO 2003). Furthermore, since these products are usually consumed semi-processed or raw, they are expected to contain higher pesticide residues than other food groups of plant origin, such as foodstuffs based on cereal processing (Shalaby and Abdou 2010; Claeys *et al.* 2011; Dragus *et al.* 2012). However, fruits and vegetables in retailers and markets may be contain pesticide residues which negatively affect consumers' health (Silipunyo *et al.* 2017; Shalaby *et al.* 2018). According to the European Union (EU 2016), a report on pesticide residues in food (with more than one pesticide) was found in 27.3% of the strawberries, peaches, apples, and lettuce samples, while 45.5% of analyzed samples (out 84,657 samples) contained quantified residues not exceeding the MRLs. Even if these substances are found at very low concentrations, long-term exposure to combinations of pesticides may have the potential to induce adverse health effects such as hormone disruption, diminished intelligence, immunity suppression, reproductive abnormalities, and cancer (Wiles *et al.* 1998; Tsatsakis *et al.* 2017; Kalliora *et al.* 2018; Ibrahim *et al.* 2018). The major route of pesticide exposure, through consumption of contaminated vegetables and fruits, continues every day (Pathak *et al.* 2013). Food monitoring and risk assessment from pesticide exposure is necessary to protect consumer health, improve the management of agricultural resources and prevent economic losses (Wolejko *et al.* 2014). The pesticide toxicity index (PTI) is a screening tool to assess the potential toxicity of multiple pesticide residues. The PTI is based on the concentration addition model to evaluate pesticide toxicity corresponding to Hazard Quotients (HQ). The HQ is used to assess ecological risk according to the guidelines of the USEPA (1998). PTI can also be used to estimate how multiple pesticide residues affect sample quality (Chaikasem and Na Roi-et 2020). Nevertheless, there are few studies on pesticide residues in the environments of Egypt, therefore, this study aimed to monitors the presence of pesticide residues in the most popularly consumed and locally cultivated vegetables, collected from major retailers and markets in Dakahlia governorate, Egypt (during 2018). Also, we checked the acquiescence of these products with maximum residue limits (MRL) and acceptable daily intake (ADI) to determine the pesticide toxicity index and health risk assessment with exposure as models of toxicity screening for food quality, safety, and sustainable agriculture.

Materials and Methods

Sampling

A total of 176 samples from 11 different vegetables, seven fruiting vegetables [i.e. tomato, cucumber, eggplant, okra, pepper, squash, and snake cucumber (qutha)]; three leafy vegetables (i.e. molokhia, watercress, and radish) and; one root vegetable(potato) were collected from major retailers and markets in Dakahlia, Egypt (Dakahlia Governorate is located east of the Delta region in northern Egypt, 30.45–31.60 N; 31.15–32.00 E), from January to April 2018. The types were selected from the most popularly consumed and locally cultivated vegetables. One kg of each sample was placed in a labeled polyethylene bag and thoroughly mixed, and prepared according to the generally recommended method (CAC 1993). Samples analyses were carried out either immediately upon their arrival to the laboratory, or they were stored at −5°C for 4 days before analysis.

Chemicals used

Sodium acetate, primary secondary amine (PSA) and magnesium sulfate were purchased from Agilent Technologies, Egypt, and Acetonitrile HPLC grade solvent was used.

Residue analysis

The QuEChERS method for pesticide residues in vegetables was followed (Lehotay *et al.* 2005; ILNAS-EN 15662:2018).The determination of residues was carried out using GC-MS/MS and LC-MS/MS after acetonitrile extraction/partitioning and clean up by dispersive SPE. The homogeneous sample was extracted under frozen conditions with the help of acetonitrile. After the addition of magnesium sulfate, sodium chloride, and buffering citrate salts (pH 5 to 5.5), the mixture was shaken intensively and centrifuged for phase separation. An aliquot of the organic phase was cleaned-upby dispersive solid-phase extraction (D-SPE) employing bulk sorbent as well as magnesium sulfate for the removal of residual water. Following clean-up with amino-sorbents (e.g. primary secondary amine sorbent, PSA) extracts were acidified by adding a small amount (less than 1 ml) of formic acid to improve the storage stability of certain base-sensitive pesticides. The final extract can be directly employed for GC- and LC-based determinative analysis. Quantification was performed using an internal standard, which was added directly before injection into the GC-MS system. The method validated 450 compounds using LCMS/MS and GC-MS/MS. The detection and confirmation of pesticide residues in the samples was madeusing GC-MS/MS and LC-MS/MS.

Apparatus

GC-MS/MS: Agilent Gas Chromatograph 7980A equipped with tandem mass spectrometer 7000B Quadrupole, EI source was used to perform analysis by using HP-5MS 5% phenyl methyl siloxane capillary column (30 m length × 0.25 mm id × 0. 25 μm film thickness). Samples were injected in a split less mode and helium was used as carrier gas (1 ml · min^{-1}). The injector temperature was 250°C, the transfer line temperature was 285°C, the ion source temperature was 280°C and the quadrupole temperature was 150°C. The oven temperature program was held at 70°C for 2 min then increased to 150°C at 25°C · min^{-1} and raised to 200°C at the rate of 3°C · min^{-1} (held for 0 min), then it went up from 200 to 280°C at 8°C · min^{-1} (held for 10 min). This resulted in a total run time of 42 min and a complete separation of all the analysts.

LC-MS/MS analysis: HPLC Agilent 1260 Series instrument was used for separation coupled to an API 6500

QtrapMS/MS. The separation was performed on a C18 column ZORBAX Eclipse XDB-C18 4.6 × 150 mm, 5 µm particle sizes (Agilent, USA). The mobile phase were as follows: Solvent A: 10 mM ammonium formate solution at pH 4 ± 0.1 in methanol-water (1:9); Solvent B: methanol. The linear gradient program was: start at 100% A; 0–13 min from 100% to 5% A; 13–21 min 5% A; 21–28 min from 5%to 100% A; 28–32 min 100% A at a flow rate of 0.3 ml· min^{-1}. The source was adjusted in the positive mode while nitrogen nebulizer, curtain, and other gas parameters were optimized according to the manufacturer's recommendations. A 400°C source temperature and 5500 V ion spray potential were common for all analytes.

Quality assurance

The criteria of quality assurance were followed to determine the performance of the standard method. The limit of quantification (LOQ) started at 0.01 mg · kg^{-1} and up depending on the pesticide type and detected module. The measurement uncertainty expressed as expanded uncertainty and in terms of relative standard deviation (at 95% confidence level) was lower than the default value set by EU (±50%). Untreated samples were fortified with the pesticide mixture and analyzed as a normal sample with a set of samples. The average recovery tests on different types of pesticides at different concentration levels varied between 70 and 120%.

Pesticide toxicity index (cumulative risk)

For each type of sample, the Toxicity Quotient (*TQ*) was computed; the *TQ* is calculated as the sum of ratios between each pesticide residue concentration and the corresponding *MRLs* (Mac Loughlin *et al.* 2018; Ramadan *et al.* 2020). The sum of *TQ* for each sample is the *PTI* as shown in the following equation (Munn *et al.* 2006; Belden *et al.* 2007):

$$TQ = \frac{C}{MRL} \text{ while } PTI = \sum TQ ,$$

where: *C* is the pesticide residue of individual pesticide concentrations (mg · kg^{-1}), *MRL* is the EU maximum residue limits (mg · kg^{-1}) (EU 2020). To address the risk of exposure to pesticide mixtures and to individual pesticides as well as to observed the degree of pesticide contamination in each sample was compared them to *MR*Ls. The *PTI* acceptable target with no risk to human health was lower than 1.00.

Health risk assessment

Health risk (*HR*) assessment of consumers caused by the intake of pesticides contaminated vegetables was described by using the health risk index (*HI*). It was calculated by dividing the estimated daily intakes (*EDI*) by their corresponding values of the acceptable daily intake (*ADI* in mg · kg^{-1}) established by WHO/FAO as shown in the following equation:

$$HI = \frac{EDI}{ADI} \times 100 .$$

The estimated daily intakes (*EDI*) of the various pesticides in each vegetable species were determined by using the following equation:

$$EDI = \frac{A \times B}{C} ,$$

where: *A*, *B*, and *C* represent the concentration of pesticide residues in vegetable (mg · kg^{-1}). The average daily intake of vegetables is shown in Table 1 (WHO 2006), and average body weight was considered to be 70.8 kg for adults (Walpole *et al.* 2012), respectively.

Table 1. Scientific names and consumption rates of analyzed vegetables in g/day based on GEMS/food total diet food balance sheet

Commodities	Scientific name	Consumption [g · day^{-1}]
Tomato	*Lycopersicon esculentum*	118.0
Potato	*Solanum tuberosum*	61.2
Cucumber	*Cucumis sativus*	5.9
Pepper	*Capsicum annuum*	13.0
Squash	*Cucurbita pepo*	11.4
Okra	*Abelmoschus esculentus*	5.3
Egg Plant	*Solanum melongena*	12.3
Snake Cucumber (Qutha)	*Cucumis melo*	5.9
Molokhia (Jews mallow)	*Corchorus olitorius*	5.3
Watercress	*Eruca sativa*	3.3
Radish	*Raphanus sativus*	3.3

Consumption rate issued by GEMS/ Food regional diet, WHO (2006)

When the *HI* is less than 100%, the food concerned is considered acceptable. If it is greater than 100%, the food concerned is considered to be a risk to the consumer (Akoto *et al.* 2016).

Results and Discussion

Eleven different vegetables corresponding to the most frequently consumed vegetables in Egypt (176 samples, 16 of each type) were collected from main retailers and markets in Dakahlia, Egypt, from January to April 2018. These vegetables included seven fruiting vegetables (tomato, cucumber, eggplant, okra, pepper, squash, and snake cucumber), three leafy vegetables (molokhia, watercress, and radish) and one root vegetable (potato).

Pesticide residues in collected vegetables

Results in Tables 2 and 3 revealed that 111 samples (63.1%) were contaminated with pesticides. Twenty three compounds detected in the contaminated samples belonged to different chemical groups (organophosphate, carbamate, pyrethroid, neonicotinoid, avermectin, etc). The obtained data showed that tomato and pepper samples were more contaminated compared than others (81.25%), followed by molokhia (75%) and cucumber (68.75%) then potato, squash, okra, and eggplant (62.5%), while watercress was the lowest (37.5%). Results in Table 2 also showed that chlorpyrifos was the most frequent compound. It was detected in 18.75% of the collected samples (33 out of 176 samples) followed by acetamiprid (9.66%) then thiamethoxam in 11 samples, while cypermethrin was the lowest detected only in one sample. According to their uses (Table 2), the majority of detected compounds were insecticides (65.2%), followed by acaricides and fungicides (13.0%), while herbicides and nematicides were 3rd (4.3%). Also, according to the WHO (2019) toxicity classification (Table 2), 12 compounds were in moderately hazardous (class II), seven pesticides belonged to class III (slightly hazardous), three compounds were in class U (unlikely to pose an acute hazard with normal use), while carbofuran is a highly toxic compound

Table 2. Pesticides detected in vegetables collected from the study area

Pesticide	LOQ [ppm]	No. of samples	% of samples	Uses	WHO classification*	Chemical group
Chlorpyrifos	10	33	18.75	insecticide	II	organophosphate
Acetamiprid	30	17	9.66	insecticide	II	neonicotinoid
Dinotefuran	25	6	3.4	insecticide	III	neonicotinoid
Indoxacarb	10	2	1.14	insecticide	II	oxadiazine
Lufenuron	10	5	2.84	insecticide	III	benzylurea
Emamectin benzoate	20	7	3.98	insecticide	II	avermectin
Chlorantraniliprole	10	3	1.7	insecticide	U	anthranilic diamides
Metalaxyl	15	3	1.7	fungicide	III	phenylamide
Thiamethoxam	15	11	6.25	insecticide	III	neonicotinoid
Profenofos	10	6	3.4	insecticide	II	organophosphate
Pendimethalin	10	2	1.14	herbicide	III	dinitroaniline
Lambda-cyhalothrin	10	3	1.7	insecticide	II	pyrethroid
Carbofuran	20	2	1.14	nematicide	Ib	carbamate
Propiconazole	20	3	1.7	fungicide	II	triazole
Hexythiazox	30	5	2.84	acaricide	U	mite growth regulator
Carbendazim	20	2	1.14	fungicide	U	benzimidazole
Cyfluthrin	15	3	1.7	insecticide	II	pyrethroid
Dicofol	30	8	4.55	acaricide	III	organochlorine
Fenpyroximate	15	8	4.55	acaricide	II	benzoate
Imidacloprid	10	2	1.14	insecticide	II	neonicotinoid
Cypermethrin	10	1	0.57	insecticide	II	pyrethroid
Carbosulfan	15	3	1.7	insecticide	II	carbamate
Malathion	10	6	3.4	insecticide	III	organophosphate

LOQ – limit of quantification; *WHO (2019) classification: Ib – highly hazardous, II – moderately hazardous, III – slightly hazardous, U – unlikely to pose as an acute hazard with normal use

Table 3. The number of analyzed vegetable samples, free, contaminated, having 1, 2 and more than two pesticides

Commodity	No. of samples	Free samples	Contaminated samples		No. of samples > *MRL*		No. of samples < *MRL*		No. of samples with one pesticide		No. of samples with two pesticides		No. of samples with more than two pesticides	
			No.	%	No.	%	No.	%	No.	%	No.	%	No.	%
Tomato	16	3	13	81.25	7	43.75	6	37.5	5	31.25	5	31.25	3	18.75
Potato	16	6	10	62.5	4	25.0	6	37.5	6	37.5	3	31.25	1	6.25
Cucumber	16	5	11	68.75	3	28.75	8	50.0	7	43.75	3	31.25	1	6.25
Pepper	16	3	13	81.25	4	25.0	9	56.25	12	75.0	0	0	1	6.25
Squash	16	6	10	62.5	3	18.75	7	43.75	10	62.5	0	0	0	0
Okra	16	6	10	62.5	3	18.75	7	43.75	9	56.25	1	6.25	0	0
Eggplant	16	6	10	62.5	2	12.5	8	50.0	8	50.0	2	12.5	0	0
Snake Cucumber	16	8	8	50.0	0	0	8	50.0	5	31.25	3	18.75	0	0
Molokhia	16	4	12	75.0	2	12.5	10	62.5	9	56.25	2	12.5	1	6.25
Watercress	16	10	6	37.5	1	6.25	5	31.25	6	37.5	0	0	0	0
Radish	16	8	8	50.0	0	0	8	50.0	7	43.75	1	6.25	0	0
Total	176	65	111		29		82		84		20		7	
%	100	36.93	63.1		16.48		46.59		47.73		11.36		3.98	

(class Ib). These data are in accordance with those obtained by Shalaby and Abdou (2020), who detected 11 pesticide residues in blood samples of occupational workers living in the same region (Dakahlia, Egypt). Most of these pesticides were moderately hazardous compounds (WHO class II). Carbofuranis a highly hazardous compound (class Ib), while chlorpyrifos was detected in 38.3% of the samples.

Tomato samples (*Lycopersicon esculentum*)

Data presented in Tables 3 and 4 revealed that 81.25% of collected tomato samples were contaminated with residues of eight pesticides, and 43.75% (7 out of 16 samples) exceeded the *MRL* established by EU (2020). Also, 31.25% of the samples had residues of one pesticide, and five samples (31.25%) had two compounds, while 18.75% was contaminated by more than two pesticides. The following pesticide residues were detected: chlorpyrifos (five samples); acetamiprid, dinotefuran, emamectin benzoate, and chlorantraniliprole (three samples) andindoxacarb, lufenuron, and metalaxyl (two samples). The *HQ*'s values of all detected compounds were below 1.00 except chlorpyrifos (2.4) and emamectin benzoate (3.5) both of which caused the risk of adverse effects following exposure to the individual pesticide.

Potato samples (*Solanum tuberosum*)

Obtained results revealed that 62.5% of potato samples had residues of six pesticides; profenofos, lambda-cyhalothrin, and propiconazole were frequented in three samples, while, thiamethoxam, pendimethalin, and carbofuran were frequented in two samples. Four out 16 potato samples (25%) had residues exceeding the *MRL*, 37.5% of the samples were contaminated with one compound, 18.75% had residues of two pesticides, while one sample (6.25%) contained residues of more than two compounds. Carbofuran had the highest *TQ* (110.0) followed by profenofos (12.0) and propiconazole (5.0), while the hazard quotient of others were below 1.00.

Cucumber (*Cucumis sativus*)

Data in Tables 3 and 5 revealed that residue of five pesticides were detected in 68.75%of collected cucumber samples. Chlorpyrifos was the most frequent (in five samples), while acetamiprid and hexythiazoxwas (four samples), dinotefuran (three samples) and carbendazim (one sample). Residues exceeding the *MRL* were found in 18.75% of the samples; while 43.75% of samples were contaminated with one pesticide, 18.75% had residues of two pesticides. One sample contained residues of more than two pesticides. However, chlorpyrifos, dinotefuran and acetamiprid may cause adverse effects on cucumber consumers because their *TQ* values were above 1.0 (4.4, 25.0 and 4.28 respectively).

Pepper (*Capsicum annuum*)

Of pepper samples 81.25% were contaminated with residues of six pesticides (13 out of 16 samples); 25% of collected samples had residues more than the *MRL*, while 75% of samples were contaminated by one pesticide (12 out 16 samples), but one sample contained residues of more than two compounds. The organophosphate insecticide, chlorpyrifos was detected in half of the collected samples (8 out 16 samples), then cyfluthrin (three samples) followed by the neonicotinoid insecticide thiamethoxam (two samples) while residues of metalaxyl, dicofol, and hexythiazox were found in one sample. The *TQ* values of detected compounds were below 1.0 except for chlorpyrifos (21.0) and dicofol (5.0).

Squash (*Cucurbita pepo*)

Results in Tables 3 and 6 indicate that 62.5% of squash samples were contaminated with residues of five pesticides; two insecticides (chlorpyrifos in three samples and acetamiprid in two samples), two acaricides (both dicofol and Fenpyroximate were found in two samples) and one fungicide (carbendazin in one sample). Residues exceeding the *MRL* were found in 18.75% of samples; all contaminated samples had the residue of one pesticide. The *TQ*'s for the individual pesticide ranged from 0.1 to 0.8 indicating no risk of adverse effects on consumers following exposure to the individual pesticides.

Okra (*Abelmoschus esculentus*)

Analysis of okra samples collected from the study region revealed that 62.5% of the samples contained residues of three insecticides, two of them belonged to organophosphate (chlorpyrifos and profenofos) and the third belonged to the neonicotinoid group (thiamethoxam); these insecticides were repeated in three samples. One insecticide contaminated 56.25% of collected samples, while one sample contained residues of two compounds. However, 18.75% of okra samples had residues exceeding the *MRL*. Profenofos hazard quotient was 1.8 which means it had hazardous effects on okra consumers.

Eggplant (*Solanum melongena*)

Overall, 37.5% of eggplant samples (six samples out of 16) had no detectable pesticide residues, and the other 62.5% contained residues of four pesticides; acetamiprid and fenpyroximate were frequented in three samples while, imidacloprid and cypermethrin were detected in two and one samples, respectively. Half of the samples contained residues at a level below the *MRL* and 12.5% had residues above the permissible limits, however, the average of detected amounts was above *ADI* (Tables 3 and 6). Half of the eggplant samples were contaminated by one pesticide, while 12.5% contained two pesticide residues. The *TQ* value of fenpyroximate fungicide was 8.5 indicating hazardous to eggplant consumers.

Snake cucumber (*Cucumis melo*)

Half of the collected samples contained detectable residues of two acaricides (dicofol and fenpyroximate were detected in five and three samples, respectively) and one insecticide (acetamiprid was repeated in three samples). Contaminated samples contained amounts below the *MRL*; the residue of one pesticide was found in 31.25%, while, 18.75% was contaminated with two compounds. The detected pesticides were of risk to consumers because their *TQ*'s been below or equal to the threshold limit (1.00).

Molokhia (*Corchorus olitorius*)

Data presented in Tables 3 and 7 showed that 75% of molokhia leaf samples had detectable pesticide residues, 12.5% (two out 16 samples) exceed the *MRL* while 62.5% contained residues that were below the permissible limits. Also, 56.25% of the samples were contaminated with one pesticide, while 12.5% were contaminated by two compounds, and 6.25% contained residues of more than two pesticides. Chlorpyrifos was the most frequently detected inmolokhia samples (nine samples), then lufenuron (three samples) while carbosulfan and emamectin benzoate were third (two samples). The average values of detected compounds were more than the *ADI*. Data also revealed that all detected pesticides had no potential health risk individually due to their *TQ* values being below or equal one.

Watercress (*Eruca sativa*)

No pesticide residues were detected in 62.5% of watercress leaf samples; but 37.5% of the samples (six out of 16 samples) had residues of malathion (three samples). Acetamiprid (two samples), and carbosulfan (one sample). Residues above the *MRL* were found in 6.25% of the samples, while 31.25%were contaminated with residues lower than the permissible limits; all contaminated samples had residues of one pesticide. The *TQ*'s of detected compounds were below or equal to one.

Radish (*Raphanus sativus*)

Overall, 50% of radish leaf samples were contaminated by residues of three insecticides; thiamethoxam was frequented in four samples, malathion and emamectin benzoate were repeated in three and two samples, respectively. The average value of detected compounds was below the *MRL* and *ADI*, and 43.75% of the samples contained residues of one insecticide while two pesticides were detected in 6.25% of samples. The *TQ* values of detected compounds ranged from 0.5 to 1.0 indicating no risk following exposure to the individual

Table 4. Residue amounts, *MRL*, *TQ*, *PTI*, *ADI*s, *EDI*s of detected pesticides and their *HI* associated with the consumption of tomato and potato samples collected from the main markets of Dakahlia Governorate

Commodity	Pesticides detected	Freq.	Rang $[\text{mg} \cdot \text{kg}^{-1}]$	Average $[\text{mg} \cdot \text{kg}^{-1}]$	*MRL* $[\text{mg} \cdot \text{kg}^{-1}]$	*ADI*s $[\text{mg} \cdot \text{kg}^{-1}]$	*EDI*s $[\text{mg} \cdot \text{kg}^{-1}]$	*HI* [%]	*HR* [Yes / No]	Half-life* [day]	*TQ***
Tomato	chlorpyrifos	5	0.01–0.37	0.24	0.1	0.01	0.0004	4.046	No	4.01	2.4
	acetamiprid	3	0.01–0.13	0.08	0.5	0.025	0.00013	0.539	No	5.69	0.16
	dinotefuran	3	0.07–0.38	0.27	0.5	NA	0.00046	NA	NA	10.06	0.54
	indoxacarb	2	0.12–0.27	0.19	0.5	0.006	0.00032	5.33	No	2.99	0.38
	lufenuron	2	0.09–0.36	0.22	0.4	0.015	0.00037	2.467	No	8.46	0.55
	emamectin benzoate	3	0.01–0.14	0.07	0.02	0.0005	0.00012	24.0	No	1.83	3.5
	chlorantraniliprole	3	0.07–0.92	0.56	0.6	1.56	0.00094	0.06	No	3.38	0.93
	metalaxyl	2	0.01–0.03	0.02	0.3	0.08	0.00003	0.0375	No	5.81	0.1
Pesticide toxicity index (*PTI*)											8.56
Potato	thiamethoxam	2	0.02–0.08	0.05	0.07	0.026	0.000043	0.168	No	3.97	0.71
	profenofos	3	0.01–0.26	0.12	0.01	0.03	0.000105	0.35	No	2.24	12.0
	pendimethalin	2	0.01	0.01	0.05	0.125	0.0000087	0.007	No	6.73	0.2
	lambda-cyhalothrin	3	0.02–0.26	0.16	0.01	0.0025	0.00014	5.6	No	2.86	0.53
	carbofuran	2	0.01–0.16	0.11	0.001	0.00015	0.000096	64.0	No	4.62	110.0
	propiconazole	3	0.01–0.07	0.05	0.01	0.04	0.000044	0.11	No	5.44	5.0
Pesticide toxicity index (*PTI*)											128.44

*half-life according to Fantke *et al*. (2014); ***TQ* – toxicity quotients, *NA* – not available, *ADI* – acceptable daily intake, *EDI* – estimated daily intake, *HI* – hazard index, *HR* – health risk, *MRL* – maximum residue limit

Table 5. Residue amounts, *MRL*, *TQ*, *PTI*, *ADI*s, *EDI*s of detected pesticides and their *HI* associated with the consumption of cucumber and pepper samples collected from the main markets of Dakahlia Governorate

Commodity	Pesticides detected	Freq.	Range $[\text{mg} \cdot \text{kg}^{-1}]$	Average $[\text{mg} \cdot \text{kg}^{-1}]$	*MRL* $[\text{mg} \cdot \text{kg}^{-1}]$	*ADI*s $[\text{mg} \cdot \text{kg}^{-1}]$	*EDI*s $[\text{mg} \cdot \text{kg}^{-1}]$	*HI* [%]	*HR* [Yes / No]	Half-life* [day]	*TQ***
Cucumber	chlorpyrifos	5	0.01–0.64	0.22	0.01	0.01	0.000019	0.185	No	4.01	4.4
	dinotefuran	3	0.06–0.16	0.11	0.5	NA	–	–	–	10.06	25.0
	acetamiprid	4	0.07–0.32	0.22	0.3	0.025	0.000019	0.074	No	5.69	4.28
	hexythiazox	4	0.01–0.04	0.02	0.5	0.03	0.0000017	0.0057	No	NA	0.04
	carbendazim	1	0.02	0.02	0.1	0.02	0.0000017	0.0085	No	5.79	0.2
Pesticide toxicity index (*PTI*)											33.92
Pepper	chlorpyrifos	8	0.01–0.46	0.21	0.01	0.01	0.00004	0.39	Yes	4.01	21.0
	cyfluthrin	3	0.02–0.26	0.11	0.3	0.01	0.00002	0.2	Yes	2.39	0.37
	thiamethoxam	2	0.04–0.12	0.08	0.7	0.026	0.00002	0.058	No	3.97	0.11
	metalaxyl	1	0.02	0.02	0.5	0.08	0.000004	0.0046	No	5.81	0.1
	dicofol	1	0.1	0.1	0.02	0.002	0.000019	0.925	Yes	3.45	5.0
	hexythiazox	1	0.02	0.02	0.5	0.03	0.000004	0.012	No	NA	0.04
Pesticide toxicity index (*PTI*)											26.62

*half-life according to Fantke *et al*. (2014); ***TQ* – toxicity quotients, *NA* – not available, *ADI* – acceptable daily intake, *EDI* – estimated daily intake, *HI* – hazard index, *HR* – health risk, *MRL* – maximum residue limit

pesticides. These results are in accordance with those obtained by Shalaby and Abdou 2020, who reported that chlorpyrifos was detected in 38.9% of blood samples collected from pesticide occupational workers in the same region (Dakahlia governorate, Egypt). This may have been becouse chlorpyrifos was the most widely used pesticide in the study area. In the same trend, data obtained by Hossain *et al*. (2015), revealed that all detected pesticide amounts exceeded the MRLs value in all vegetable samples collected from Savar bazaar, Bangladesh, except forchlorpyrifos in tomato and lady's finger, and cypermethrin in tomato and eggplant, respectively. The presence of pesticides in vegetables makes these vegetables more hazardous

Table 6. Residue amounts, *MRL*, *TQ*, *PTI*, *ADI*s, *EDI*s of detected pesticides and their *HI* associated with the consumption of squash, okra, eggplant, and snake cucumber samples collected from the main markets of Dakahlia Governorate

Commodity	Pesticides detected	Freq.	Range [mg · kg^{-1}]	Average [mg · kg^{-1}]	*MRL* [mg · kg^{-1}]	*ADI*s [mg · kg^{-1}]	*EDI*s [mg · kg^{-1}]	*HI* [%]	*HR* [Yes / No]	Half-life* [day]	TQ**
Squash	chlorpyrifos	3	0.01–0.18	0.13	0.01	0.01	0.000021	0.21	No	4.01	0.65
	acetamiprid	2	0.02–0.12	0.07	0.2	0.025	0.000011	0.044	No	5.69	0.35
	carbendazim	1	0.01	0.01	0.1	0.02	0.000002	0.0082	No	5.79	0.1
	dicofol	2	0.01–0.16	0.08	0.02	0.002	0.000013	0.65	No	3.45	0.8
	fenpyroximate	2	0.02–0.3	0.16	0.08	0.01	0.000026	0.26	No	1.51	0.8
Pesticide toxicity index (*PTI*)											2.7
Okra	chlorpyrifos	3	0.02–1.1	0.44	0.01	0.01	0.000033	0.33	No	4.01	0.88
	profenofos	3	0.03–0.13	0.09	0.1	0.03	0.0000068	0.023	No	2.24	1.8
	thiamethoxam	3	0.02–0.09	0.06	0.01	0.026	0.0000045	0.0173	No	3.97	0.009
Pesticide toxicity index (*PTI*)											2.689
Eggplant	acetamiprid	3	0.02–0.24	0.16	0.2	0.025	0.000028	0.112	No	5.69	0.27
	fenpyroximate	3	0.01–0.26	0.17	0.3	0.01	0.00003	0.3	No	1.51	8.5
	imidacloprid	2	0.06–0.12	0.09	0.6	0.06	0.000016	0.026	No	3.7	0.15
	cypermethrin	1	0.2	0.2	0.5	0.00125	0.000035	2.8	No	4.24	0.28
Pesticide toxicity index (*PTI*)											9.2
Snake Cucumber	acetamiprid	3	0.03–0.14	0.1	0.3	0.025	0.0000084	0.0336	No	5.69	0.33
	dicofol	5	0.01–0.02	0.02	0.02	0.002	0.0000017	0.085	No	3.45	1.0
	fenpyroximate	3	0.01–0.04	0.02	0.08	0.01	0.0000017	0.017	No	1.51	0.25
Pesticide toxicity index (*PTI*)											1.58

*half-life according to Fantke *et al*. (2014); **TQ – toxicity quotients, *NA* – not available, *ADI* – acceptable daily intake, *EDI* – estimated daily intake, *HI* – hazard index, *HR* – health risk, *MRL* – maximum residue limit

Table 7. Residue amounts, *MRL*, *TQ*, *PTI*, *ADI*s, *EDI*s of detected pesticides and their HI associated with the consumption of molokhia, watercress, and radish samples collected from the main markets of Dakahlia Governorate

Commodity	Pesticides detected	Freq.	Range [mg · kg^{-1}]	Average [mg · kg^{-1}]	*MRL* [mg · kg^{-1}]	*ADI*s [mg · kg^{-1}]	*EDI*s [mg · kg^{-1}]	*HI* [%]	*HR* [Yes / No]	Half-life* [day]	TQ**
Molokhia (Jews mallow)	chlorpyrifos	9	0.01– 1.4	0.23	0.01	0.01	0.000017	0.174	No	4.01	0.46
	lufenuron	3	0.01- 0.02	0.015	0.01	0.015	0.000001	0.0073	No	8.46	0.75
	carbosulfan	2	0.01–0.02	0.015	0.002	0.005	0.000001	0.022	No	2.11	0.75
	emamectin benzoate	2	0.01	0.01	0.01	0.005	0.0000008	0.0152	No	1.83	1.0
Pesticide toxicity index (*PTI*)											2.96
Watercress	malathion	3	0.01–0.04	0.03	0.02	0.03	0.0000014	0.0047	No	2.48	0.6
	acetamiprid	2	0.01	0.01	0.01	0.025	0.00000047	0.0019	No	5.69	1.0
	carbosulfan	1	0.01	0.01	0.002	0.005	0.00000047	0.0094	No	2.11	1.0
Pesticide toxicity index (*PTI*)											2.6
Radish	malathion	3	0.02–0.2	0.1	0.02	0.03	0.0000047	0.017	No	2.48	0.5
	thiamethoxam	4	0.01	0.01	0.01	0.026	0.00000047	0.0018	No	3.97	1.0
	emamectin benzoate	2	0.01	0.01	0.01	0.005	0.0000004	0.008	No	1.83	1.0
Pesticide toxicity index (*PTI*)											2.5

*half-life according to Fantke *et al*. (2014); **TQ – toxicity quotients, *NA* – not available, *ADI* – acceptable daily intake, *EDI* – estimated daily intake, *HI* – hazard index, *HR* – health risk, *MRL* – maximum residue limit

to consumers. Analysis of 20 vegetable samples collected from the River basin area (Thailand) by Chaikasem and Na Roi-et (2020) revealed that 95% of the samples were contaminated with organophosphate insecticides followed by pyrethroid (40%) and organochlorine (20%).

Pesticide toxicity index (PTI) values (cumulative risk)

A hazard quotient (*HQ*) [= Toxicity Quotients, (*TQ*)] for a single pesticide, while pesticide toxicity index (*PTI*) for a mixture of pesticides; and *PTI* is the sum HQs of the pesticides in a mixture (Goumenou and Tsatsakis 2019). Data on the behavior in food vegetables is a key aspect of current risk and impact assessment. This is because consumers' exposure to pesticides is generally caused by residues in crops grown for human consumption (Fantke and Juraske 2013; Malhat *et al.* 2018; Chaikasem and Na Roi-et 2020). Data in Tables 3–6 show the cumulative risks posed by the consumption of vegetables by humans. Also revealed that the pesticide toxicity index (*PTI*) amounts were >1.00 from all contaminated samples (63.1% of collected samples). The highest *PTI* (128.44) was noticed in potato samples followed by cucumber (33.92), and pepper (30.16); while the *PTI* values of eggplant, tomato, molokhia, squash, okra, watercress, and radish samples were 9.2, 8.56, 2.96, 2.7, 2.689, 2.6 and 2.5, respectively, but snake cucumber had the lowest value (1.58). These results agree with those obtained by Chaikasem and Na Roi-et (2020) who reported that the pesticide risk values for vegetables collected from the River Basin area (Thailand) were >1.0 and therefore raise concern to the consumer. These data indicated that pesticide residues in collected samples could accumulate with time, and this may be the cause of adverse chronic effects on humans in the study period. There is a critical need to survey the pesticide residues in order to standardize the amount of pesticide application. Also, Khan *et al.* (2020) reported that the pesticides bifenthrin, amamectin, and difenoconazole which were detected in fruits and vegetables collected from some agricultural areas of Lahore (Pakistan) pose a significant health risk to consumers because of their health risk indices. Also, Hassain *et al.* (2015), reported that the highest health indices were noticed for ethion (5.7) in lady's finger, carbaryl (1.09) in tomato, carbofuran (1.17) and chlorpyrifos (1.97) were detected in brinjal samples collected in Bangladesh.

Health risk assessment

When assessing chronic exposure, the level of pesticide exposure over a lifetime and the likely effects on health from such exposure is considered (Gad Alla *et al.* 2013). This assessment method is well developed and considers the mean levels of exposure in relation to the *ADI* values established for individual pesticides. In the case of consumers exposed to residues of chronically toxic pesticides, their health would only be at risk if their dietary intake exceeded the *ADI* every day for an extended period of time. The estimated acceptable daily intake (*EDI*) was calculated separately for each pesticide in an exposure assessment. If the acceptable daily intake (*ADI*) was not exceeded in any commodity, a chronic consumer risk can be excluded. Data in Table 4 revealed that the hazard index (*H*I) for contaminated tomato samples ranged from 0.375% formetalaxyl to 24.0% of the *ADI* for emamectin benzoate, while its values ranged from 0.007% for pendimethalin to 64% for carbofuran in potato samples.Data in Table 5 revealed that the hazard index (*HI*) values ranged from 0.0057% for hexythiazox to 0.185% for chlorpyrifos in cucumber samples, but it ranged from 0.0046 to 0.925% for metalaxyl and dicofol, respectively in pepper samples. In squash samples, *HI* values ranged from 0.0082 to 0.65% for carbendazim and dicofol (Table 6), but it ranged from 0.0173% for thiamethoxam to 0.33% for chlorpyrifos in okra samples. Also, in eggplant samples, the values of *HI* ranged from 0.026% for imidacloprid to 2.8% for cypermethrin, while it ranged from 0.017% to 0.085% for fenpyroximate and dicofol, respectively in snake cucumber. In molokhia, watercress, and radish sample, the hazard index did not exceed the *ADI*; its amounts ranged from 0.0073 to 0.174% for lufenuron and chlorpyrifos; and ranged from 0.0019 to 0.0094% for acetamiprid and carbosulfan; from 0.0018 to 0.017% for thiamethoxan and malathion (Table 7). It is clear that from the obtained data of risk assessment of pesticide residues in the studied vegetables no risk will be found from consuming these vegetables. These data are in accordance with those obtained by Akoto *et al.* (2016) who reported that when the *HI* is less than 100%, the food concerned is considered acceptable. If it is greater than 100%, the food concerned is considered to be a risk to the consumer. These data also indicated thatcarbofuran had the highest *HI* value (64%) in potato tubers. Since the *EDI* did not exceed the *ADI*: this means that all detected pesticides had no potential health risk individually due to the *HI* values (below the cut-off limit of 100). While Hossain *et al.* (2015) reported that the single and cumulative health risk assessment associated with pesticide exposure in vegetables, showed negligible effects on consumers, but pesticide exposure by vegetable consumption, can be considered as a concern due to the consumption of uncookedfoods such as carrots, lettuce, cucumbers if they are unwashed. Also, Seo *et al.* (2013) reported that pesticide residues detected in dried vegetables collected from Seoul, Korea, had a negligible risk associated with exposure.

Pesticide half-life values

Half-life is the time required for half of the pesticide to break down (Bajwa and Sandhu 2014). According to Fantke *et al.* (2014), the half-life of pesticide values in this study ranged from 1.51 days for fenpyroximate (acaricide) to 10.06 days for dinotefuran (insecticide); the highest half-life values (dinotefuran) were detected in tomato and cucumber fruits, followed by lufenuron (8.46 days) which was detected in tomato fruits and molokhia leaves, then pendimethalin (6.73 days) was detected in potato tubers, while fenpyroximate was detected in squash, eggplant and snake cucumber (quatha). It is therefore important that steps are taken to minimize pesticide residues in food. Thorough washing of vegetables and cooking could help to eliminate these residues (Tables 4–7).

Conclusions

Cumulative risk assessment is a very important approach to access hazards due to multiple residues. The estimation of dietary exposure is an essential requirement. The lack of consumption data for different groups of the population is considered to be one of the barriers to conducting this study. So, in developing countries such as Egypt, some legislative measures should be taken to survey the pesticide residues in foodstuff because of potential health hazards and to make the products safe for consumers. Obtained data revealed that there was no risk of adverse effects following exposure to individual pesticides; while the combined risk index of pesticide residues was a significant health risk for consumers according to the individual risk index. Therefore, this study concluded that there is a need for strict regulation and regular monitoring of pesticide residues in foodstuff for consumers' health protection.

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