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Assessment of pesticide residues in vegetables selected from different Egyptian governorates

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Abstract

This study aimed to assess the levels of contamination by pesticide residues in several types of vegetables collected from different regions in Egypt. A total of 100 samples of vegetables (pepper, tomato, cucumber, and strawberry) were collected from markets in five cities (Al-Obour, Al-Salheia El-Gadida, Giza, Zagazig, and Fayed) and analyzed for the presence of 42 different pesticide residues. The Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) method was used to extract the target pesticides, which were then quantified using Gas Chromatography-Mass Spectrometry (GC-MS/MS) and Liquid Chromatography-Mass Spectrometry (LC-MS/MS) techniques. The results showed that 72% of the vegetable samples contained detectable levels of pesticide residues, with 21% exceeding the European Union Maximum Residue Levels (EU-MRLs) and 51% containing residues below the MRLs. The detected residues were primarily insecticides (56.4%) and fungicides (43.6%), with tomato and strawberry samples showing the highest frequency of both types of pesticides. Tomato also had the highest absolute intake from consumption (2.89 g/kg BW/day), followed by strawberries, peppers, and cucumbers (0.47, 0.159, and 0.096 g/kg BW/day, respectively). A hazard index (HI) was used to assess the dietary risk posed by the pesticide residues, with tomato having the highest contribution value. These findings highlight the need for Integrated Pest Management (IPM) programs to reduce the excessive use of pesticides, particularly in relation to raw food commodities. Action is required to minimize the unacceptable risks identified in this study.

Keywords: Food safety, Pesticide residues, Risk assessment, Estimated daily intake, Monitoring

Introduction

Fruit and vegetables have been a cornerstone of healthy dietary recommendations. They have potential health-promoting effects beyond providing basic nutrition needs in humans, including their role in reducing inflammation and their potential preventive effects on various chronic disease states such as cardiovascular disease and cancer leading to premature mortality decreasing years loss of individuals' life and years-to-come lived with disability/morbidity. Consumers are now choosing fruits and vegetables not only for their content of vitamins, minerals and fiber, but also for their concentration of dietary bioactive with its anti-inflammatory effects [1]. In agriculture, pesticides are considered a quick, and easy solution for controlling weeds and insect pests, improving production and productivity of agriculture commodity to feed the ever growing population, controlling vector borne disease like malaria and reducing the resultant mortality and morbidity. Surprisingly; the global consumption of pesticides is about two million tons per year and out of which 45% is used by Europe alone, 25% is consumed in the USA, and 25% in the rest of the world. Despite their benefits, pesticides can be hazardous to humans and environment and non-intended organisms ranging from beneficial soil microorganisms to insects, plants, fish and birds. Environmental contamination or prevailing use of pesticides can expose the general population to pesticide residues leading to serious and prolonged toxicity. It was estimated that a minimum of 300,000 people die from pesticide poisoning each year, with 99% of them from low- and middle-income countries in 2009 [2].

Pesticides in food are monitored by the Environmental Protection Agency (EPA) and the Food and Drug Administration (FDA). One of the top priorities in securing and preserving community and public health, is food safety. Food safety is particularly important to ensure the healthiness of food, especially to fruits and vegetables as they are consumed in substantial and considerable quantities customarily without any processing. They are susceptible to pests at any point in the production chain, from the field through storage ahead till food consumption. Their pesticide residues are yet present in the vegetable-treated products, which may constitute a potential hazards for consumers. Some of these hazards were identified in fruits and vegetables were because of incorrect application of pesticides either by the producers' application or the insufficient monitoring of the contaminated soil and/or water [3]. Major causes of the environment pollution were from prejudicial human activities and improper application, spillage, and decomposition. However and despite all precautions, a very minute amount of pesticide-residues can remain in the treated crop. The Maximum Residue Level (MRL), the maximum quantity of residue that is legally permitted on a food material, ensures that both imported and exported goods are moderately safe to consume. Pesticide misuse, false positives from naturally occurring compounds, variances in national MRLs standards, a lack of registered pesticides, and improper pesticide application can all cause MRLs to be exceeded [4].

The sources of the MRLs, however, can affect the frequency of infractions. However, the creation of MRLs is based on information from Supervised Trials Mean Residues (STMR), Acceptable Daily Intake (ADI), Acute Reference Dose (ARfD), as well as data from Good Agricultural Practice (GAP) that has been registered nationally. The percentages of violation will be very different, for instance, because the maximum permissible residue of profenofos on tomatoes is 10 mg/kg in the Codex and 0.01 mg/kg in European standards. In order to determine the actual risk of exposure, it is crucial to compare the findings with a more reliable toxicological endpoint, such as ADI or ARfD. The Egyptian ought to take into account creating MRLs based on regional best practices for agriculture and locally administered paths. The major tool for ensuring that the pesticides were applied in assembly with good agricultural practices is the monitoring program. These programs when applied in conformity with Good Agricultural Practices, treated goods should not have exceeded levels of pesticide residue [5].

The health risks of pesticides are regularly evaluated through monitoring programs for EU nations. According to the yearly DG SANCO report, 47% of the fruits, vegetables, and grains eaten in Europe in 2004 had pesticide residues [6]. Pesticides that can be used in certain foods and feed commodities have maximum residual limitations, or tolerances, determined by the Environmental Protection Agency (EPA). These restrictions are put in place to safeguard people from hazardous pesticide levels in their food [7]. A variety of pesticides, including organochlorine, organophosphorus, carbamate, insecticides, fungicides, and herbicides, are used by farmers all over the world to prevent the devastating crop loss that can result from pests and diseases as well as to boost agricultural productivity to ensure a sufficient supply of food for the expanding population [8].

Food contamination creates severe health issues worldwide, ranging from minor ailments to fatal ones [9]. Therefore; it is well established that contaminated food poses a risk to the general public health. However; producing, processing, moving, and handling food can all lead to food contamination [10; 11; 9; 12]. There are two types of pollution: short-term pollution at high concentrations of chemicals (induced by inadvertent release or contamination from the source) and long-term pollution of low concentrations of chemicals (produced by the progressive diffusion of pollutants in food) [13]. Different chemical classes or families typically produce dissimilar symptoms. The amount of pollutants in food ingested in relation to the daily amount of food can be used to estimating the level of pollutants in the human body [14].

Numerous studies have demonstrated that persistent organic pollutants, such as organochlorine insecticides, have a variety of negative impacts, including aberrant immune system development, birth abnormalities, and foetal death [15; 16].

Because of this, pesticides are regarded as one of the world's top environmental and health risks [17]. Many nations and international organizations, including the European Union, the World Health Organization, and the United Nations Environment Program, have acclaimed that both organic and inorganic pollutants pose a serious risk to health, particularly the health of children [18]. They have gradually released a number of suggestions or guidelines intended to limit or outlaw the use of these pollutants or pollutant products. For instance, the amount of lead in the environment has decreased as a result of several countries banning the addition of tetra-methyl lead to gasoline [19]. For consumers protection from exposure to unacceptable levels of pesticide residues in food and feed, the European Commission has set maximum residue levels (MRLs), defined as the highest possible level of a pesticide residue that is legally authorized in food and feed. Based on the results obtained from environmental sample analysis, it has been proven that the Modified QuEChERS method coupled to Gas Chromatography GC-MS/MS with electron capture detector (GC-ECD) which are analytical procedures for routine analysis and simultaneous determination of selected electronegative pesticides in fruits and vegetables with high water content. These procedures are suitable not only for fruits and vegetables with high water content, but also for samples containing large amounts of pigments and dyes [3].

For risk assessment analyses that could be linked to accidental intakes of contaminants at very high levels and could have severe unfavorable effects on the human body, quantitative data on the concentration of contaminants in food are an essential tool [20] . Though pesticides assist increase crop productivity, the amount and variety of food consumed, and likewise the development of some diseases [21]. Pesticides can be categorized according to their unique biological activity or their target species, such as fungicides, herbicides, insecticides, and acaricides, in addition to functional groups [22; 23]. The maximum daily consumption that a person is permitted to consume during their lifetime without posing a significant risk to that person has been determined by numerous health and environmental protection agencies as "acceptable daily intake" (ADI) levels. Environmental pollution is, without any doubts, a serious global concern. Many nations have made positive efforts to limit the use of pesticides. Procedures and approaches are utilized to evaluate the detrimental impacts brought on by pollutants as a result of pollutant risk assessment. To evaluate past, present, and future exposure to any environmental pollutants, a risk assessment can be carried out. The majority of the time, analyses of material hazards are based on scientific research on the activities, exposure, quantity, and toxicity of chemicals. The amount of pollutants present in the environment, food, and/or products; the number of people exposed to the pollutants; and the damages of pollutants all affect the risks [24].

Many organochlorine pesticides have been outlawed or have had their usage severely restricted in Europe and North America, yet they are still sold and used in Africa. Residential pesticide use in Egypt is high. Adolescents in Egypt are exposed to pesticides through non-occupational and para-occupational pathways. In addition to the hormonal and physiological changes associated with puberty, there are also significant developmental changes in the brain, primarily the prefrontal cortex. The impact of environmental exposures can vary across developmental periods and consequences of prolonged exposure can last into adulthood [25]. The purpose of this study was to assess the levels of a group of pesticide residues in the commonly consumed vegetables in Egypt, and to evaluate their health risk according to estimated quantity of exposure.

Material and methods

Samples

A total of 100 vegetable samples (pepper, tomato, cucumber, and strawberry) were collected from local markets of five Egyptian cities: Al-Obour (Qalyubia Governorate), Al-Salheia El-Gadida (Sharqia Governorate), Giza (Giza Governorate), Zagazig Sharqia (Governorate), and Fayed (Ismailia Governorate)). Each representative vegetable sample was made up of 10 identical commodity subsamples that were simultaneously obtained from each market using random sampling. Vegetable samples were packed in proper bags and stored at 4^{o}c until analysis.

Sample preparation and analysis

About 2 kg of each vegetable sample (pepper, tomato, cucumber, and strawberry) was thoroughly washed with tap water, chopped and blended using a waring laboratory blender. Each sample was chopped and ground in accordance with the generally suggested procedure described by the Codex Alimentarius Commission in 1993 for no more than two days prior to analysis.

According to Anastassiades *et al.,* [26] the QuEChERS method was used to extract pesticides from the vegetable samples. A 50 ml polypropylene (PP) tube containing 10 gm of each sample was weighed, 10 ml of acetonitrile was added, and the tube was forcefully shaken for one minute. Phase separation was achieved by centrifuging the liquid at 4000 rpm for 5 min after adding buffering citrate salts (pH 5 to 5.5), containing 4g of magnesium sulphate, and 1g of sodium chloride. For analysis, an aliquot of the organic phase was directly loaded into LC-MS/MS. Dispersive solid phase extraction (DSPE). Extracts from the samples were evaporated and then redissolved along with injection standard for GC-MS/MS analysis after cleaning with primary secondary amine sorbent (PSA). Aldrin was used as an internal standard for quantification, and it was added to the GC-MS/MS system right before injection. GC MS/MS and LC-MS/MS were used for the identification and confirmation of pesticide residues in the samples.

Calculation of ADI and HI

Comparing the established acceptable daily intake (ADI) with the estimated acceptable daily intake (EDI), which is based on the concentration of pesticide residues and food consumption, gives the risk assessment. The EDI (mg/kg BW/day) for each pesticide residue that was violated was computed by multiplying the mean pesticide residue concentration (mg/kg) x food consumption and then dividing by the typical adult's body weight (60 kg) of each commodity. Based on GEMS/FOODS from the WHO's Global Environment Monitoring System [27], acceptable daily intake was determined.

$$EDI = \frac{Concentration\ of\ pesticideresidue \times Food\ consumed}{Body\ weight} \tag{1}$$

The daily consumption rate of vegetables was derived conclusively for this study from the reports of WHO/ FAO [28], WHO/ Global Environment Monitoring System-Food Contamination, Monitoring and Evaluation Program average consumption cluster diets [27], and Gad Alla et al., [29]. If data from food balance sheets are unavailable for a commodity, the consumption level for a comparable food is used (WHO 1997). Because there isn't a strawberry consumption rate available, the consumption level of a comparable food is used. The scientific names and daily intake rate (g/day) for the used vegetable samples are given in **Table 1** . The EU Pesticides Database served as the source for both the maximum residue limits (MRLs) and the established acceptable daily intake (ADI) values. Using the health risk index, the health risk for consumers from consuming pesticide-contaminated samples was described (HI). It is calculated by dividing the Estimated Daily Intake (EDI) by the corresponding values of the Acceptable Daily Intake (ADI in mg/kg) specified by WHO/FAO as stated in the equation 2:

$$HI = \frac{EDI}{ADI}100 \tag{2}$$

Table 1. Scientific names and consumption rate of studied commodities in g/day based on GEMS/food total diet food balance sheet. Consumption rate (g/day) based on WHO/Global Environment Monitoring SystemFood Contamination, Monitoring and Assessment Program average consumption cluster C diets[27].

Common Name	Scientific Name	Family Name	Crop type	Consumption (g/day)*
Cucumber	*Cucumis sativas L.*	Cucurbitaceae	Vegetable	5.9
Pepper	*Capsicum Annum L.*	Solanaceae	Vegetable	13
Strawberry	*Fragaria ananassa L.*	Rosaceae	Vegetable	20
Tomato	*Solanum lycopersicum*	Solanaceae	Vegetable	118

According to the European Food Safety Authority (EFSA) [30; 31] El-Sawy et al. and [32]. When the HI is less than 100%, the food concerned is considered acceptable. If it is above 100%, the food concerned is considered a risk to the consumer [33].

Results and Discussion

Use of excessive pesticides contaminates soil, water and finally enters the food chain and contaminates the food produced. The International Agency for Research on Cancer has found sufficient evidence of carcinogenic potential in most of the pesticides beyond the threshold limit. The United Nations Environment Program estimates accidental pesticide poisoning causing 20, 000 deaths and 1 million cases of illness per year worldwide [34]. Forty-two commonly used pesticides in agriculture were identified in this study. The broad scope analyzed includes numerous groups of pesticides such as organophosphorous, organo–chlorine, pyrethroids and other groups of pesticides widely used or outlawed in Egypt. According to a directive issues by Egypt's Agriculture Pesticides Committee (Codex+EU), pesticide residue levels should be compared to Codex Alimentarious when it is available and to EU-MRLs when Codex MRLs are not accessible. In this investigation, only the codex Alimentarious MRLs and the Agriculture Pesticides Committee decree were used to compare the monitoring data. **Table 2** listed the number of samples evaluated, the range of pesticides found, the average in mg/kg, the number of chemicals violated in the samples analyzed, and the status of each pesticide/commodity combination in the registration system established by the Agricultural pesticide committee (APC). A number of 32 out a total of 42 pesticides were detected in strawberry fruits. Fluopyram had the highest pesticide concentration in the samples, whereas Iprodion had the lowest pesticide residue (**Table 2**).

Data revealed that 13 (52%) of the strawberry fruit samples had no detectable pesticide residues. Whereas, a total of 12 samples (48%) contained pesticide residues, of which 10% were contaminated samples and contained residues at levels below the MRLs, and 8% had residues over the allowed limits (**Table 2**). However, according to the APC regulation, the breach was found in 8% of cases when comparing the results to (codex + EU restrictions). Bifenazate, methamidophos, fluopyram, metalax, captan, propargite, and pyrimethanill are seven pesticide residues that recorded greater amounts than their regulated EU MRL values (**Table 2** and **Figure 1**). The discovered quantities of pyrimethanill contamination (LOQ to 0.076, average 0.048 mg/kg) were not too far from the MRL (0.05 mg/kg). Otherwise, the residues of both fluopyram and methamidophos exhibited a serious issue because their concentrations in strawberries exceeded their MRL values by 4 and 9 fold, respectively. This indicates that it is necessary to regulate their use.

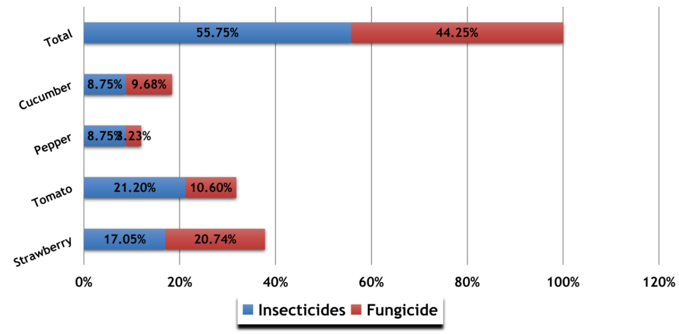

Figure 1. The percentage of detected pesticide residues in samples based on pesticide type.

Regarding the outcomes of the tomato samples, which were the second crop of the vegetables under study, roughly 17 pesticides were detected in the tomato samples (25), as shown in **Table 3**. Among the pesticides found, only 8 (bifenazate, captan, fluopyram, chlorfenapyr, chlorpyrifos, dimethoate, lambda-cyhalothrin, and thiofanat-methyle) had concentrations greater than the MRLs reported for tomato (**Table 2** and **Figure 2**). However, data in **Table 3** revealed that 28% (7) of tomato fruit samples had no detectable pesticide residues (25). While pesticide residues were discovered in 18 samples (72%) of which 48% (12 samples) were contaminated tests recorded residual amounts comparable to the worldwide MRLs for them in tomato and 24% had residues over the allowed levels, pesticide residues were also discovered in other samples.

Fenarimol fungicide, followed by Malathion insecticide, had the highest pesticide mean in samples, whereas Fenpropathrin and Metalaxyl had the lowest pesticide residue levels (**Table 4**). However, pesticide residues were discovered in 21 samples (84%), of which 36% (9 samples) had residues over the allowed limits and 48% (12 samples) of contaminated samples had residues at levels higher than the MRLs (**Table 4**). According to the decision of the Agriculture Pesticides Committee in Egypt, a violation was found in one sample when results were compared to codex and EU limitations, and in the other eight samples when findings were compared to solely EU limits. Data in **Table 5** regarding cucumber fruits showed that the majority of the investigated pesticides (12 out of 42) were detected in cucumber samples. Only 2 insecticides (Abamectin & Acetamiprid) and 2 fungicides (Captan & Penconazole) out of the detected 12 pesticides had concentrations greater than the MRLs indicated for cucumber, as shown in **Table 5**.

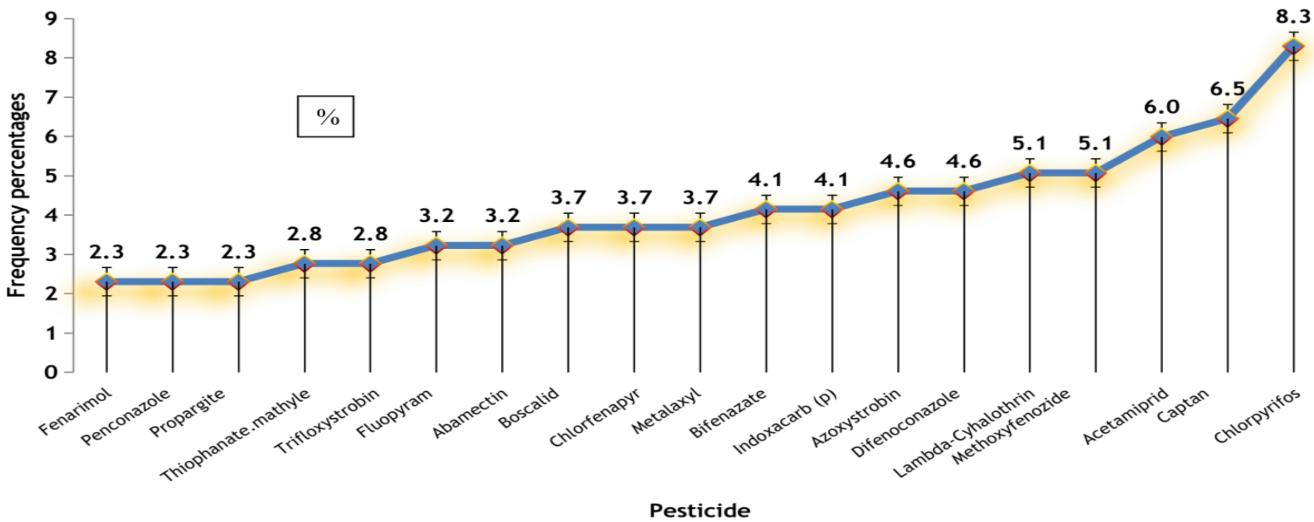

Figure 2. Frequency percentages of most detected pesticide residues in vegetable samples.

In total, 16% of the cucumber samples had no detectable pesticide residues, while the remaining 84% contained detectable residues which 68% of contaminated samples contain resides at levels lower than the MRL's and 16 % (4 samples) had residues above the permissible limits as shown in **Figure 3**. The MRLs are often set well below the thresholds deemed safe for humans. It is important to understand that MRLs are not safety limits; food residues might have levels beyond MRLs while still being safe to eat [35]. According to IFOAM [36], MRLs are not a guarantee of "zero health risk" in this situation; rather, they are only indicators of whether or not Good Agricultural Practices (GAP) are being violated, not a sign of a health danger. Risk exposure should therefore be assessed using toxicological endpoints like Acceptable Daily Intake (ADI). When it comes to earlier Egyptian investigations, the pattern of pesticides in tomato indicated that the detection levels of dimethoate, pirimiphos-methyl, and profenofos were 0.461, 0.114, and 0.28, respectively [37].

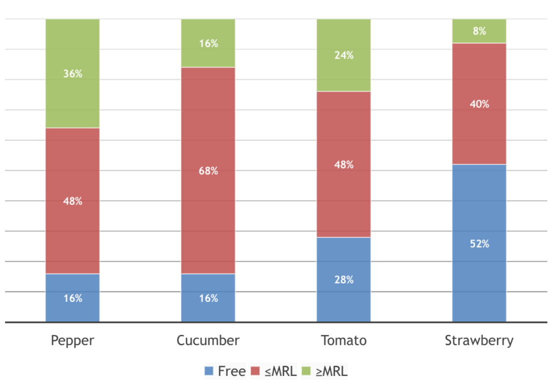

Figure 3. The contamination and the violation percentages per each commodity of vegetables.

Otherwise, greater residual levels of profenofos and Malathion were found in tomato samples taken from various areas in Egypt, according to Dogheim *et al.,* [38]. Additionally, greater profenofos residue levels were found in strawberry samples taken from eight local markets in six Egyptian governorates [39; 40; 38], but not in tomato or strawberry samples taken for this study. The organophosphorus pesticides (thiometon, phorate, and chlorpyrifos-methyl) were found in cucumbers in a different investigation by Mansour *et al.,* [41] for the monitoring of pesticides and heavy metals. The levels of pesticides in some vegetables gathered from neighborhood markets in Cairo governorate were assessed by Farag *et al.,* [42]. According to their findings, strawberries had the highest levels of contamination with various pesticides, with mean contamination levels of 0.034, 0.023, 0.033, 0.024, and 0.050 mg/kg, respectively, for ethion, propargite, permethrin, profenofos, and chlorpyrifos. Pepper was found to contain only two different types of insecticides (sulfur, methomyl). Pesticides were not found in tomatoes or cucumbers. Additionally, Ibrahim *et al.,* [43] assessed the pesticide residues in certain vegetables purchased at local marketplaces in eight governorates around Egypt. They came to the conclusion that the reported negative samples for pepper and cucumber were 19.4% and 27.9%, respectively. For pepper and cucumber, respectively, the recorded positive samples were 80.6% and 72.1%. According to Badr *et al.,* [44] , who assessed the pesticide residues in Egyptian crops, profenophos measured pesticide levels in tomato and cucumber at 0.56 and 0.28 mg/kg, respectively. The levels of pesticide residue found in vegetable samples by Loutfy *et al.,* [45] and Ahmed *et al.,* [46] are consistent with our findings.

According to Dogheim *et al.,* [38] , the relatively limited amount of pesticides used in the research areas and the widespread awareness and usage of integrated pest management (IPM) programs may be responsible for this low contamination level. Con-

Table 2. Pesticide residues levels found in strawberry fruits, frequencies, their corresponding MRLs, number of violated samples and the status of registration of each detected pesticide in analyzed samples collected from different governorates during 2020.

Active Ingredient	Freq.	Residues (mg/kg)			MRL		VC	EAPC
		Min	Max	Mean	Codex	EU	(>MRL)	
Acetamiprid	1	0.039	0.039	0.039	0.5	0.3		R-NRI
Azoxystrobin	7	0.012	0.12	0.054	10	2		R-NRI
Bifenazate	4	0.012	0.25	0.083	2	0.1	1	R-RI
Bifenazole	3	0.01	0.029	0.016	-	0.1		NR
Boscalid	6	<LOQ	0.11	0.043	-	0.9		R-NRI
Buprofezin	1	0.081	0.081	0.081	3	3		NR
Captan	5	0.017	0.294	0.133	15	0.1	2	R-RI
Carbendazim	2	0.02	0.07	0.045	0.5	0.1		Banned, 2018
Chlorantraniliprole	1	<LOQ	<LOQ	<LOQ	0.3	1		R-NRI
Chlorpyrifos	3	<LOQ	<LOQ	<LOQ	0.3	0.01		R-NRI
Cyproconazole	1	0.017	0.017	0.017	0.01	0.05		NR
Difenoconazole	4	<LOQ	0.042	0.031	2	1		R-NRI
Fluopyram	2	0.031	0.44	0.236	0.4	0.1	1*	R-RI
Flutazole	1	<LOQ	<LOQ	<LOQ	NA	0		NR
Flutriafol	1	0.021	0.021	0.021	1.5	1.5		R-NRI
Imidacloprid	2	<LOQ	<LOQ	<LOQ	0.5	0.5		R-NRI
Indoxacarb (p)	3	0.013	0.06	0.024	0.02	0.6		R-NRI
Iprodion	2	0.012	0.013	0.013	10	0.05		Banned, 2018
Lambda-Cyhalothrin	2	<LOQ	<LOQ	<LOQ	0.02	0.02		R-NRI
Metalax	3	0.01	0.09	0.033	0.5	0.05	1	R-NRI
Methamidophos	3	0.068	0.097	0.078	0.01	0.01	3*	NR, 2007
Methoxyfenozide	3	0.038	0.076	0.059	2	2		R-NRI
Paclobuttazol	1	<IOQ	<IOQ	<IOQ	NA	0.01		NR
Penconazole	1	0.018	0.018	0.018	0.5	0.1		R-NRI
Pirimicarb	1	0.11	0.11	0.11	-	0.5		R-NRI
Propargite	5	<LOQ	0.095	0.067	2	0.05	2	NR
Propiconazole	2	0.01	0.046	0.028	3	0.05		NR
Pyraclostrobin	4	0.01	0.053	0.022	1.5	0.1		R-RI
Pyrimethanil	3	<LOQ	0.076	0.048	0.1	0.05	2	R-RI
Spirodiclofen	2	<LOQ	0.025	0.013	2	0.5		R-NRI
Tetraconazole	1	<LOQ	<LOQ	<LOQ	NA	0.2		R-NRI
Thiophanate.mathyle	2	0.038	0.068	0.053	NA	0.1		R-RI

* MRL of EU+ codex Alimentarious, others were of EU, Freq= Number of Pesticides Found on the Commodity, R= Registered, R1= Recommended, NR= Not Registered, NR1= Not Recommended, EAPC=Egyptian Agriculture Pesticide Committee , VC=Violated Compound

Table 3. Pesticide residues levels found in tomato, frequencies, their corresponding MRLs, number of violated samples and the status of registration of each detected pesticide in analyzed samples collected from different governorates during 2020.

Active Ingredient	Freq.	Residues (mg/kg)			MRL		VC	EAPC
		Min	Max	Mean	Codex	EU	(>MRL)	
Abamectin	1	0.010	0.010	0.010	0.05	0.09	-	R-RI
Acetamiprid	3	0.010	0.010	0.010	0.2	0.5	-	R-RI
Azoxystrobin	3	0.014	0.022	0.015	3	3	-	R-RI
Bifenazate	5	<LOQ	0.16	0.160	0.5 (2007)	0.1	2	R-RI
Boscalid	2	0.010	0.030	0.020	X	3	-	R-NRI
Captan	4	0.050	0.368	0.228	5 (2008)	0.1	2	R-NRI
Chlorfenapyr	7	0.010	0.071	0.022	0.4 (2019)	0.01	5	R-RI
Chlorpyrifos	5	0.021	0.053	0.036	1	0.01	5	R-RI
Cyfluthrin	1	<LOQ	<LOQ	<LOQ	0.2 (2008)	0.05	-	NR
Difenoconazole	3	<LOQ	<LOQ	<LOQ	0.6	2	-	R-RI
Dimethoate	3	0.010	0.020	0.017	NA	0.01	2	R-NRI
Fluopyram	2	0.100	0.410	0.255	0.5	0.1	1*	R-NRI
Indoxacarb	6	0.010	0.040	0.022	0.5	0.5	-	R-RI
Lambda-Cyhalothrin	7	<LOQ	0.110	0.060	NA	0.01	2	R-NRI
Methoxyfenozide	7	0.010	0.062	0.024	2 (2005)	2	-	R-RI
Thiofanat-methyle	4	0.080	1.200	0.570	X	1	1	R-NRI
Trifloxystrobin	6	<LOQ	0.010	0.010	0.7 (2006)	0.01*	-	R

* MRL of EU+ codex Alimentarious, others were of EU, Freq= Number of Pesticides Found on the Commodity, R= Registered, R1= Recommended, NR= Not Registered, NR1= Not Recommended, EAPC=Egyptian Agriculture Pesticide Committee , VC= Violated Compound

Table 4. Pesticide residues levels found in pepper , frequencies, their corresponding MRLs, number of violated samples and the status of registration of each detected pesticide in analyzed samples collected from different governorates during 2020.

Active Ingredient	Freq.	Residues (mg/kg)			MRL		VC	EAPC
		Min	Max	Mean	Codex	EU	(>MRL)	
Chlorofenapyr	1	0.01	0.01	0.01	0.05	0.01	-	R-NRI
Chloropyrifos	7	0.01	0.07	0.03	2	0.02	5	R-NRI
Dimethoate	1	0.03	0.03	0.03	0.5	0.01	-	R-NRI
Fenarimol	5	<LOQ	0.3	0.22	0.5	0.1	3	R-NRI
Fenpropathrin	1	<LOQ	<LOQ	<LOQ	1	0.01	-	R-NRI
Fluopyram	2	0.03	0.05	0.04	3	0.1	-	R-NRI
Imidacloprid	1	0.09	0.09	0.09	1	0.5	-	R-NRI
Malathion	1	0.2	0.2	0.2	0.1	0.01	1*	R-NRI
Metalaxyl	2	<LOQ	<LOQ	<LOQ	1	1	-	R-NRI
Methoxyfenozide	1	0.06	0.06	0.06	2	2	-	R-NRI
Spinetoram	4	0.01	0.07	0.04	0.4	0.5	-	R-RI

* MRL of EU+ codex Alimentarious, others were of EU, Freq= Number of Pesticides Found on the Commodity, EAPC=Egyptian Agriculture Pesticide Committee , VC=Violated Compound

Table 5. Pesticide residues levels found in cucumber , frequencies, their corresponding MRLs, number of violated samples and the status of registration of each detected pesticide in analyzed samples collected from different governorates during 2020.

Active Ingredient	Freq.	Residues (mg/kg)			MRL		VC	EAPC
		Min	Max	Mean	Codex	EU	(>MRL)	
Abamectin	6	0.01	0.07	0.04	0.03	0.04	2*	R-RI
Acetamiprid	9	0.06	0.1	0.12	0.3	0.3	1*	R-RI
Captan	5	<LOQ	0.7	0.21	3	0.1	2	R-NRI
Chloropyrifos	3	0.03	0.04	0.03	0.05	0.05	-	R-NRI
Cypermethrin	2	0.17	0.2	0.18	0.2	0.2	-	R-NRI
Difenoconazole	3	<LOQ	<LOQ	<LOQ	0.2	0.2	-	R-RI
Fenpropathrin	1	0.03	0.03	0.03	0.5	0.5	-	R-NRI
Fluopyram	1	0.07	0.07	0.07	0.5	0.1	-	R-NRI
L-Cyhalothrin	2	0.03	0.04	0.03	0.1	0.1	-	R-NRI
Malathion	1	0.01	0.01	0.01	0.2	0.02	-	R-RI
Metalaxyl	3	0.16	0.3	0.23	0.5	0.5	-	R-RI
Penconazole	4	0.01	0.04	0.02	0.06	0.01	2	R-NRI

* MRL of EU+ codex Alimentarious, others were of EU, Freq= Number of Pesticides Found on the Commodity, R= Registered, R1= Recommended, NR= Not Registered, NR1= Not Recommended , EAPC=Egyptian Agriculture Pesticide Committee , VC=Violated Compound

trarily, numerous pesticides, including those forbidden in Egypt, are nevertheless found in a variety of environmental components, including groundwater, surface water, fished, mussels, medical plants, soils, and sediments [47; 48; 49; 50; 46]. Additionally, some pesticides were found in only one crop, as shown in the corresponding Tables for the crops of strawberry (13 pesticides), tomato (2, cyfluthrin & trifloxystrobin), and pepper (2, fenarimol & spinetoram), despite the fact that many common pesticides were found in multiple crops in some of the samples used for the current investigation. In light of these findings, a distinction between the types of pesticide residue found on the Egyptian samples during the current investigation and those conducted previously was noted. The dominant pesticide group was adjusted in addition to the pesticide itself. A regular pesticide mentoring survey should therefore be recommended because it is related to the evaluation of the safety of fruits used in human food. The Ministry of Agriculture's various control programs, which vary from one field and/or location to another depending on the type of injury and the different environmental conditions, may be to blame for the high incidence of the majority of tested pesticides in some vegetable samples of the current investigation.

Pesticide frequencies (Detected pesticides type)

According to **Figure 1**, the types of pesticide residues that were found in vegetable samples were insecticides and fungicides, with percentages of 55.75% and 44.25%, respectively. Insecticides were found in descending order at frequencies of 21.2, 17.05, 8.75, and 8.75% in tomatoes, strawberries, cucumbers and peppers. In descending order, fungicides with rates of 20.74, 10.60, 9.68, and 3.23% were found in strawberries, tomatoes, cucumbers, and peppers. But compared to pepper and cucumber, strawberry and tomato had the highest frequencies of both insecticides and fungicides. These findings contrasted with those of Ozowicka *et al.,* [51] who discovered that fungicides were used four times as frequently as insecticides, and were generally consistent with finding of Gad Alla *et al.,* [52] . In addition, **Figure 2** lists the nineteen (19) pesticides that were found most frequently in Egyptian vegetables examined in 2020, listed in order of frequency percentages: Chlorpyrifos (8.3%), Captan (6.5%), Acetamiprid (6.0%), Lambda-Cyhalothrin (5.1%), Methoxyfenozide (5.1%), Azoxystrobin (4.6%), Difenoconazole (4.6%), Bifenazate (4.1%), Indoxacarb (4.1%), Boscalid (3.7%), Chlorfenapyr (3.7%), Metalaxyl (3.7%), Abamectin (3.2%), Fluopyram (3.2%), Thiophanate.mathyle (2.8%), Trifloxystrobin (2.8 %), Penconazole (2.3%), Propargite (2.3%) and Fenarimol (2.3%), in addition to 23 other pesticides (11 insecticides, 9 fungicides, 2 acaricides and 1 herbicide) with a frequently number of 1-4 with a percentage < 2% (**Table 6**). This may indicate a more intense use of insecticides than fungicides, which is inconsistent with pest control behavior in fruit and vegetable, due to fungal diseases that are expected to infect fruits.. However, the usage of numerous types and vast quantities of pesticides caused insects to become resistant, which led to the need of more pesticides overall.

Detected pesticides groups

The detected pesticides could be categorized into a variety of groups, as shown by **Table 6** and **Figure 4**, with the most frequent groups being organophosphorus OPs, followed by DMI-fungicide, Strobilurin, Neonicotinoids, pyrethroids, Pyridine carboxamides, Phthalimides, and Diacylhydrazines, with frequencies percentages of 12.44%, 9.68%, 9.22%, 7.37%, 7.3 The most often identified pesticide class in Egyptian vegetables in 2020 was OP's (12.44%), which has been the case for a number of years. The majority of OPs were found in vegetable samples from 2011 that were previously collected (41.5%; Sohair *et al.,* [29]); they were also found in fruit samples from 2007 that were previously collected (29.4%; Gadallah *et al.,* [52]) and they are widely utilized in Egypt. The effectiveness and low cost may be the causes of the product's prolonged use in Egyptian markets, and farmers find it difficult to change their usage patterns. Concerns are raised by a review conducted by Roshini *et al.,* [53] that exposure to OP pesticides at levels currently regarded as acceptable may have had a negative impact on human reproductive function and survival. Contrarily, OPs are hazardous to the nervous system and have been substantially eliminated from agriculture in several nations during the past ten years [52]. However, they are not outlawed and continue to be used on some food crops, according to the EWG's 2012 list of the "Dirty Dozen" endocrine disruptor pesticides (The-dirty-dozen-eco-group, 2012). Fungicides containing strobilurin have been used extensively in agricultural fields for many years. The FRAC group 11 fungicides known as strobilurin or QoI are very effective at controlling a variety of common vegetable diseases [54] . Stabilurins'

Table 6. The detected pesticide frequencies in analyzed Egyptian vegetable and fruit samples, 2020.

SN	Substance classified*	Chemical Name	Type	Freq. No
1	Organophosphate	Chlorpyrifos	Ins.	18
2		Dimethoate	Ins.	4
3		Malathion	Ins.	2
4		Methamidophos	Ins.	3
5	Pyrethroid	Cyfluthrin	Ins.	1
6		Cypermethrin	Ins.	2
7		Fenpropathrin	Ins.	2
8		L-Cyhalothrin	Ins.	11
9	DMI**	Cyproconazole	Herb.	1
10		Difenoconazole	Fung.	10
11		Flutazole	Fung.	1
12		Flutriafol	Fung.	1
13		Penconazole	Fung.	5
14		Propiconazole	Fung.	2
15		Tetraconazole	Fung.	1
16	Strobilurin	Azoxystrobin	Fung.	10
17		Pyraclostrobin	Fung.	4
18		Trifloxystrobin	Fung.	6
19	Pyridinecarboxamides	Boscalid	Fung.	8
20		Fluopyram	Fung.	7
21	Neonicotinoids	Acetamiprid	Ins.	13
22		Imidacloprid	Ins.	3
23	Benzimidazole	Carbendazim	Fung.	2
24		Thiophanate.M	Fung.	6
25	Carbamates	Pirimicarb	Ins.	1
26	Growth regulator	Paclobuttazol	Fung.	1
27		Buprofezin	Ins.	1
28	Avermectins	Abamectin	Ins.	7
29	Acramite	Bifenazate	Acar,	9
30	Anilinopyrimidines	Pyrimethanil	Fung.	3
31	Arylepyrrole	Chlorfenapyr	Mite	8
32	Diacylhydrazines	Methoxyfenozide	Ins.	11
33	Diamides	Chlorantraniliprole	Ins.	1
34	Dicarboximides	Iprodion	Fung.	2
35	Imidazole	Bifenazole	Acar.	3
36	Oxadiazine pesticide	Indoxacarb (p)	Ins.	9
37	Phenylamide :acrylalanin	Metalaxyl	Fung.	8
38	Phthalimides	Captan	Fung.	14
39	Pyrimidines	Fenarimol	Fung.	5
40	Spinosyns	Spinetoram	Ins.	4
41	Sulfite ester	Propargite	Acar.	5
42	Tetronic acids	Spirodiclofen	Acar.	2
Total				217

*-Types; Acar, Acarecides-Ins, insecticide- Fung, fungicide -Herb, herbicide, -Substance classified is referred to: PAN pesticide database, - Freq; The total number of frequencies for the positive samples of all vegetable under study, **DMI: Demethylation Inhibitors

unique, non-target-specific fungicidal activity. Previously, strobilurins were thought to be less hazardous to mammals [55], but a number of authors have noted that this is not entirely true due to gaps in the toxicological endpoints of fungicides [56]. These pesticides are intended to control fungi, but because of their broad-spectrum method of action, they can also have unintended consequences. Therefore, Strobilurin toxicity may result in ecosystem imbalance and food-web disturbance.

The development of synthetic pyrethroids involved extensive chemical modifications that made them more toxic and less biodegradable in the environment [57; 58]. Despite this, pyrethroids still have negative effects because they can lead to endocrine disruption, liver function impairment, and respiratory problems when exposed to them over an extended period of time.

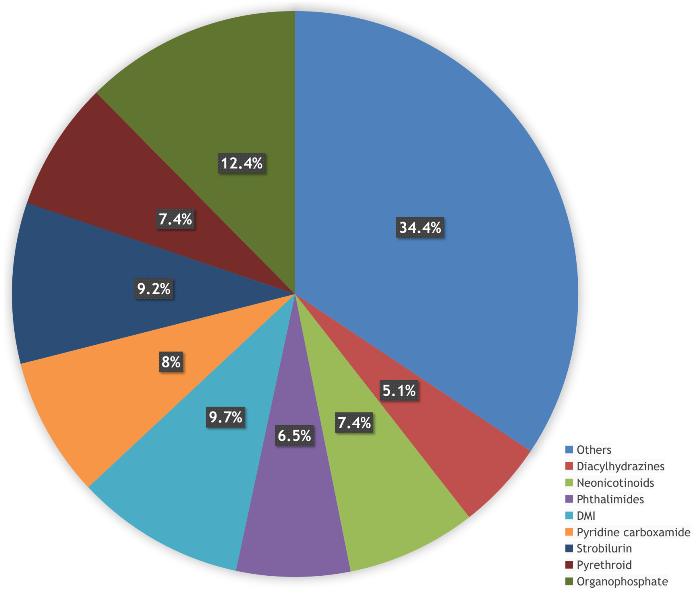

Figure 4. The most detected pesticides groups in vegetable samples analysed during 2020.

Health risk assessment

Because of the potentially harmful effects, dietary exposure to pesticides raises health concerns. Food quality and safety must be ensured in order to reduce the increased health risks associated with eating foods that contain pesticide residues. In order to reduce the presence of and exposure to pesticide residue, especially non-authorized pesticides, monitoring programs for the detection of pesticide residues in food should give enhanced health risk estimations for hygienic activities. **Table 0.4.3** clarifies approved daily intake (ADI) and estimated daily intake (EDI) for pesticides. Strawberry, pepper, and cucumber had the lowest absolute intakes from ingesting the commodity, with 0.47, 0.159, and 0.096 g/kg b.w/day, respectively. Tomato had the highest absolute intakes, with 2.89 g/kg BW/day (**Table 0.4.3**). Once more, to evaluate chronic exposure, the amount of pesticide exposure throughout a lifetime and any potential health impacts of

Table 7. Pesticide estimated daily intake (EDI), acceptable daily intake (ADI) and hazard index (HI%).

Commodity	Estimated dietary intake (1EDI, (g/kg b.w/day)					2ADI		HI
Pesticide	Strawberry	Tomato	Pepper	Cucumber	Total	(g /kg BW/day)	(source; year)	
Bifenazate	0.028	0.315	-	-	0.342	10	JMPR-2006 & EFSA-2017	3.424
Fluopyram	0.079	0.502	0.009	0.007	0.596	10	JMPR-2010	5.956
Dimethoate	-	0.033	0.007	-	0.040	2	JMPR-1995	1.997
Abamectin	-	0.02	-	0.004	0.023	1	JMPR-2015	2.327
Thiophanate.mathyle	0.018	1.121	-	-	1.139	30	JMPR-2013	3.796
Difenoconazole	0.01	0.01	-	0	0.020	10	JMPR-2007, 2010, 2013	0.205
Chlorpyrifos	0.002	0.07	0.007	0.003	0.082	10	JMPR-2009	0.819
Cypermethrin	-	-	-	0.018	0.018	5	EFSA, 2018	0.362
Propargite	0.022	-	-	-	0.022	10	JMPR-1999	0.222
Boscalid	0.014	0.039	-	-	0.054	40	JMPR-2006	0.134
Indoxacarb	0.008	0.043	-	-	0.051	10	JMPR-2005	0.507
Others	0.29	0.738	0.136	0.064	1.226			3.6606

1EDI: Estimated daily intake based on vegetable consumption data shown in Table a.
ADI: Acceptable daily intake, HI: Hazard index= (EDI/ADI*100)
JMPR is the Joint FAO/WHO Meetings on Pesticide Residues and EFSA is European Food Safety Authority.
**taking into consideration that in the absence of ADI, the value of 10 g /kg BW/day is usually set by default

such exposure must be taken into account [29]. This assessment method, which was carefully developed, considers average exposure levels in relation to the ADI values determined for specific pesticides. The estimated daily intake (EDI) for each pesticide was det independently in an exposure assessment. If consumers are exposed to chronic hazardous pesticide residues, their health won't be at risk unless they consume more food than the ADI each day for a long time.

Hazard index

According to data in **Table 8**, the hazard index (HI) for contaminated strawberry samples ranged from 0.0001% for Chlorantraniliprole to 0.785% of the ADI for fluopyram, while its values ranged from 0.015% for Azoxystrobin to 5.015% for fluopyram in tomato samples, ranged from 0.0013% (Metalaxyl) to 0.479% (Fenarimol) in pepper samples, but ranged from 0.0003 to 0. The data from the risk assessment of pesticide residues in the tested commodities show that there is no risk associated with eating these vegetable. These findings are consistent with those made by Akoto *et al.,* [33] , who stated that when the HI value is less than 100%, the concerned commodity is regarded acceptable. If the HI value is larger than 100%, the concerned food is considered to pose a risk to consumers. According to recent statistics, Fluopyram has a tomato HI value of 5.015 percent, which was the highest. Due to the HI values, all tested pesticides had no individual potential health harm because the EDI did not exceed (below the cut-off limit of 100). According to Hossain *et al.,* [59] , pesticide exposure in vegetables has minimal effects on consumers both individually and collectively, although it is possible that eating raw foods like cucumbers that haven't been washed could increase the risk of pesticide exposure.

Additionally, Seo *et al.,* [60] revealed that the danger of exposure to pesticide residues found in dried vegetables gathered from Seoul, Korea, was minimal. The current findings demonstrated that there is no link between Egyptian consumers' long-term exposure to pesticide residues from eating raw vegetable and health risks. It should be noted that the current study is restricted to a select few vegetable. Furthermore, rather than assessing the cumulative exposure to several pesticide residues in crops, the predicted risk assessment via long-term exposure is based on toxicological evaluation of the individual chemicals.

Contributed pesticides in total hazard index

In order of contribution percentages (via consumption of a single commodity) in the analyzed Egyptian vegetables during 2020, **Figure 5** lists the 13 high contributed pesticides, which include eight insecticides, one acaricide, and four fungicides. In order of decreasing contribution to the HI, the following pesticides were used: Fluopyram (25.4%), thiophanate-mathyle (16.2%), Bifenazate (14.6%), Abamectin (9.9%), Dimethoate (8.5%), Chlorpyrifos (3.5%), Methamidophos (2.8%), Lambda-Cyhalothrin (2.6%), Captan (2.2%), Indoxacarb (2.2%), and Fenarimol (2.0%), Cypermethrin (1.5%) and Buprofezin (1.3%) in descending order, while each of the remaining 29 pesticides contributed less than 1% of the HI.

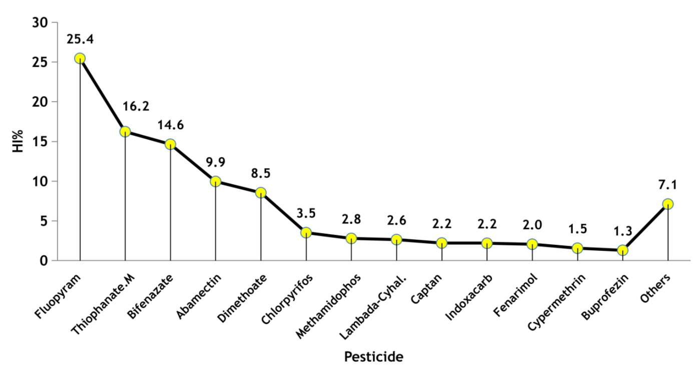

Figure 5. The most contributed pesticides in total hazard index (HI%).

Contributed commodities in total hazard index

The calculated contributions of each commodity to the HIs are shown in **Figure 6**, along with the commodities that make

Table 8. The estimated intake (EDI) and the hazard index (HI) of pesticide residues in Egyptian vegetable samples.

Pesticides detected	Strawberry		Tomato		Pepper		Cucumber	
	EDI	HI	EDI	HI	EDI	HI	EDI	HI
Abamectin	-	-	0.02	1.97	-	-	0.0036	0.361
Acetamiprid	0.013	0.019	0.02	0.03	-	-	0.0118	0.017
Azoxystrobin	0.018	0.009	0.03	0.02	-	-	-	-
Bifenazate	0.028	0.278	0.31	3.15	-	-	-	-
Bifenazole	0.005	0.054	-	-	-	-	-	-
Boscalid	0.014	0.036	0.04	0.1	-	-	-	-
Buprofezin	0.027	0.3	-	-	-	-	-	-
Captan	0.044	0.044	0.45	0.45	-	-	0.0204	0.02
Carbendazim	0.015	0.05	-	-	-	-	-	-
Chlorantraniliprole	0.002	0.0001	-	-	-	-	-	-
Chlorfenapyr	-	-	0.04	0.15	0.002	0.01	-	-
Chlorpyrifos	0.002	0.017	0.07	0.7	0.007	0.07	0.0031	0.031
Cyfluthrin	-	-	0.01	0.02	-	-	-	-
Cypermethrin	-	-	-	-	-	-	0.0181	0.362
Cyproconazole	0.006	0.028	-	-	-	-	-	-
Difenoconazole	0.01	0.102	0.01	0.1	-	-	0.0005	0.005
Dimethoate	-	-	0.03	1.67	0.007	0.33	-	-
Fenarimol	-	-	-	-	0.048	0.48	-	-
Fenpropathrin	-	-	-	-	0.001	0.003	0.003	0.01
Fluopyram	0.079	0.785	0.5	5.02	0.009	0.09	0.0069	0.069
Flutazole	0.002	0.017	-	-	-	-	-	-
Flutriafol	0.007	0.07	-	-	-	-	-	-
Imidacloprid	0.002	0.003	-	-	0.02	0.03	-	-
Indoxacarb (p)	0.008	0.081	0.04	0.43	-	-	-	-
Iprodion	0.004	0.007	-	-	-	-	-	-
Lambda-Cyhalothrin	0.002	0.008	0.12	0.59	-	-	0.0033	0.017
Malathion	-	-	-	-	0.043	0.01	0.001	0.0003
Metalaxyl	0.011	0.014	-	-	0.001	0.00125	0.0223	0.028
Methamidophos	0.026	0.65	-	-	-	-	-	-
Methoxyfenozide	0.02	0.02	0.05	0.05	0.013	0.01	-	-
Paclobuttazol	0.002	0.008	-	-	-	-	-	-
Penconazole	0.006	0.02	-	-	-	-	0.002	0.007
Pirimicarb	0.037	0.183	-	-	-	-	-	-
Propargite	0.022	0.222	-	-	-	-	-	-
Propiconazole	0.009	0.013	-	-	-	-	-	-
Pyraclostrobin	0.007	0.024	-	-	-	-	-	-
Pyrimethanil	0.016	0.008	-	-	-	-	-	-
Spinetoram	-	-	-	-	0.008	0.02	-	-
Spirodiclofen	0.004	0.043	-	-	-	-	-	-
Tetraconazole	0.002	0.042	-	-	-	-	-	-
Thiophanate.mathyle	0.018	0.059	1.12	3.74	-	-	-	-
Trifloxystrobin	-	-	0.02	0.05	-	-	-	-

EDI: Estimated daily intake, ADI: Acceptable daily intake, HI: Hazard index = (EDI/ADI*100) JMPR is the Joint FAO/WHO Meetings on Pesticide Residues and EFSA is European Food Safety Authority.**taking into consideration that in the absence of ADI, the value of 10 g /kg BW/day is usually set by default

up the majority of those contributions. Tomatoes were the commodities that contributed the greatest to hazard (HI). As a result, they posed some risks to consumers and had exposures that were above acceptable limits, but generally, home processing methods including washing, heating, and frying may lower the levels of residues in vegetables.

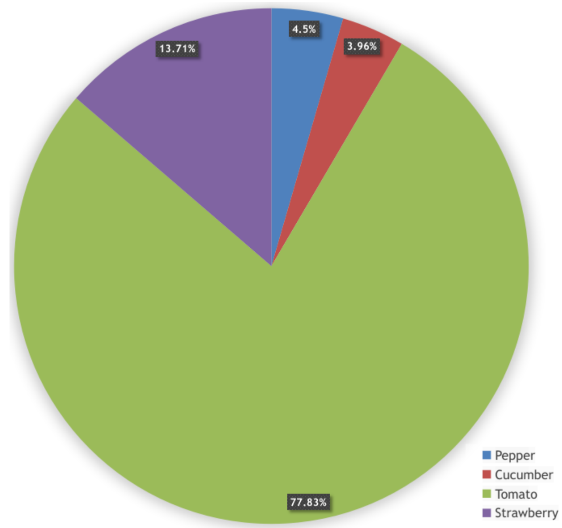

Figure 6. The most contributed commodities in total hazard index (HI %).

Conclusion

With the exception of levels of several insecticides, such as Lambada-cyhalothrin (tomato), Methamidophos (strawberry), and Malathion (pepper), most pesticide residues were present in the most frequently consumed foods at below detectable levels, and those present above detection levels were in trace amounts. Generally speaking, coordinated efforts are required to improve food quality and safety as well as to manage pesticide residues in both the environment and humans. The presence of prohibited pesticides in local markets needs to be addressed by the government authorities. Implementing IPM programs correctly within the GAP framework, creating new pesticide application methods, and shifting toward the production of organic foods will all assist sustainable development. Further studies should be conducted to provide further evidence on the long-term health effects of pesticide exposure through diet.

Abbreviations

ADI: Acceptable daily intake
BW: Body weight
CCCF: Codex Committee on Contaminants in Foods
CIFOCOss: Chronic Individual Food Consumption Database Summary statistics
EFSA: European Food Safety Authority

FAO:	Food and Agriculture Organization of the United Nations
GC:	Gas chromatography
	Global Environment Monitoring System
GEMS/Food:	Food Contamination Monitoring and Assessment Programme
IPCS:	International Programme on Chemical Safety
ISO:	International Organization for Standardization
IUPAC:	International Union of Pure and Applied Chemistry
JECFA:	Joint FAO/WHO Expert Committee on Food Additives
JECFA:	Joint FAO/WHO Expert Committee on Food Additives
JMPR::	Joint FAO/WHO Meeting on Pesticide Residues
LOD:	limit of detection
LOQ:	limit of quantification
MS:	Mass spectrometry
MS/MS:	Tandem mass spectrometry
QuEChERS:	Quick, Easy, Cheap, Effective, Rugged, and Safe
USA:	United States of America
WHO:	World Health Organization

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