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Monitoring of pesticide residues in some Egyptian herbs, fruits and vegetables

[1]Farag, R.S., [1]Abdel Latif, M.S., [2]Abd El-Gawad, A.E. and [2]Dogheim, S.M.

[1]*Biochemistry Department, Faculty of Agriculture, Cairo University, Egypt*
[2]*Agricultural Research Center, Ministry of Agricultural, Dokki, Egypt*

Abstract: One hundred thirty two samples of fruits, vegetables, herbs and spices collected from Egyptian local markets were analyzed for pesticide residues. Contamination with pesticide residues reached 54.55% while samples free from contamination reached 45.45%. Only one sample from 132 analyzed samples violated the Maximum Residue Limits (MRLs) of the Codex Committee. From the 132 analyzed samples, 72 samples (54.55%) were contaminated, from which 43.18% contaminated with residues from one pesticide residue, 6.06% with 2 residues and 5.3% with more than 2 residues. In addition, 2 caraway and one fennel samples contained 4 pesticide residues, one sample of marjoram contained 5 pesticide residues and one mint sample contained 6 pesticide residues. Six of the pesticides detected as residues in the analyzed food items were considered to be carcinogens at different levels of assurance.

Keywords: Pesticide recidue, Egyptian herbs, Egyptian fruits, Egyptian vegetables

Introduction

Pesticide residues in food have historically lagged far behind many comparable hazards as a cause for public health concern and action (Correia *et al*., 2000; Eskenazi *et al*., 2008). Pesticide residue contaminating food is the problem focused worldwide because of its direct implications on human health and international trade (Sanborn *et al*., 2004). Reliable residue analysis data resulting from monitoring programs in foods, even if limited, may be of great value indicating the possible risks of pesticide exposure on human health and on international trade (DAF and FSAI, 2006).

The Maximum Residue Limits (MRLs) as food standards differ widely for the same pesticide on the same commodity between countries as well as with the international Codex Committee standards (Codex, 2010). However, scientists cannot say for sure that there is ever a "safe" level of pesticide residues in food because many of the chemical messengers in our bodies function at precisely minute quantities of ppm or even ppb (Boobis, *et al*., 2008).

Consumer protection is very highly considered by governments and authorities responsible for pesticides registration and use in each country and by the international organizations. Risk issues that should be focused and studied are cumulative risk assessment (cocktail effect), endocrine disruption and carcinogenicity (PAN, 2002). Pesticide residue monitoring data in food serve in evaluating and clarifying the situation of potential human risk and trade problems. Such data could help decision makers in reviewing and reconsidering the registration and use of pesticides in the country. The aim of this study was to investigate a simple limited monitoring program for use of pesticide residue data in assessment of the possible risks that might affect human health and international trade.

Materials and Methods

Sampling

One hundred thirty two samples representing herbs [fennel, anise, basil, caraway, chamomile, marjoram, dill, mint and hibiscus (n=40)], vegetables [strawberry, onion, cucumber, lettuce, okra, pepper, peas, green beans and tomatoes (n=54)] and fruits [dates, orange and lemon (n=38)] were randomly collected from the local markets at Cairo. All herbs, vegetables and fruits were maintained at 2-5°C until analysis.

Extraction

For fruit and vegetable samples, a known weight of the food commodity sample (25 g) was mixed with a known concentration of pesticide internal standard (Aldrin and Ditalimphos). The mixture was thoroughly homogenized with acetone for 3 min then celite was added (10 g) to the mixture and homogenized again for 1 min. The homogenate was filtered, transferred to a separatory funnel containing sodium chloride (20 g) and vigorously shaken for 3 min. Dichloromethane (25 ml) was added, shaken for 3 min and allowed to stand for 30 min. The lower

*Corresponding author.
Email: *radwanfarag@yahoo.com*

aqueous phase was discarded. The organic phase was concentrated near dryness, dissolved in ethyl acetate: cyclohexane mixture (10 mL 1:1, v/v) and filtered through a membrane filter (0.45 µm). For the herb samples, extraction was carried out as reported by the DFG Multiresidue Method S 19 (1987 and 1992) using accelerated solvent extractor (ASE) apparatus.

The cleanup step was carried out as reported by the DFG Multiresidue Method S 19 (1987 and 1992) using a gel permission chromatography (GPC) cleanup for Gas Chromatography (GCs) and silica gel cleanup column for GC-ECD.

Determination of pesticide residues

All analytical methods and instruments were fully validated as a part of the laboratory quality assurance system and were audited and accredited by the United Kingdom Accreditation Service (UKAS) according to requirements of ISO 17025. The criteria of quality assurance were described by Dogheim *et al*. (2002). The recoveries were between 70-120%.

Organochlorine pesticides, synthetic pyrethroids and other compounds containing halogen atoms, nitro groups and other electronegative groups were determined by GC- ECD. Phosphorus- and sulfur-containing compounds were determined by GC-FPD. Phosphorus- and nitrogen-containing compounds were determined by GC- NPD. GC/MS and LC/MS/MS were used for the analysis of pesticide residues which can not detected by ECD, FPD or NPD. The qualitative and quantitative analysis of pesticides residues were performed under the following conditions:

Gas Chromatography, "Agilent 7890 A series" equipped with different detectors, i.e., electron capture (ECD), flame photometry (FPD) and nitrogen phosphorus (NP) detectors were used. Analysis of the pesticides was performed on two capillary columns, i.e., HP-5 (5%-Phenyl-methylpolysiloxane) and DB-35 (35%-Phenyl-methylpolysiloxane). The dimensions of each column were 30 m length x 0.25 mm inner diameter and coated with 0.25 µm film thickness of the stationary phase. Nitrogen was used as a carrier gas at a flow rate of 1ml/min.

Gas Chromatography, "Perkin Elmer, Clarus 600 series" equipped with MSD (Mass spectroscopy detector). Analysis of the pesticides was performed on a capillary column, Elite-5 [Dimethyl polysiloxane (5% diphenyl)], the dimensions of the column were 30 m length x 0.25 mm inner diameter and coated with 0.25 µm film thickness of the stationary phase. Helium was used as a carrier gas at a flow rate of 1ml/min.

The temperatures of injector and interface were 250°C and 300°C, respectively. The temperature program for all GCs was as follows; initial temperature was 100°C for 1min, raised at rate of 25°C/min to 170°C, isothermal for 1 min, raised at a rate of 3°C/min to 230°C, then isothermal for 1 min, finally raised at a rate of 8°C/min to 300°C, then isothermal for 5 min.

Liquid chromatography, "Agilent 1200A series" equipped with triple quadruple 6410 was used for pesticide residues determination. The analysis was performed using 150 mm x 4.6 mm stainless steel column packed with ZORBAX SB-Phenyl 600 Bar (1.8 µm film thickness). The column temperature was maintained at 25°C. The mobile phase was consisted of acetonitrile (ACN) and water both with HCOOH (0.1%) and its flow rate was 0.6 ml/min. The gradient elution program was as follows; initial, 10% ACN linear to 28 min., then 98% ACN to 30 min, 100% ACN to 31 min 10% ACN to 45 min.

Calculation

The pesticide residue concentration was deduced from the following equation:

$$C=\frac{a}{b}\times d\times f$$

Where:

C= Concentration of pesticide residue (ppm).

A= The concentration of the identified analyte in the sample solution from GCs and LC/MS/MS determination step (ppm).

B= The sample equivalent in the extraction step (ppm).

D= The dilution factor of GPC cleanup step.

F= The dilution factor of Silica gel column cleanup step.

Results

Residues of two hundred and forty one pesticides were analyzed using the multi residue method (S19) in 132 samples of different food commodities available in the local markets in Great Cairo, Egypt. Herbs (fennel, anise, basil, caraway, chamomile, marjoram, dill, mint and hibiscus), vegetables (onion, cucumber, lettuce, okra, pepper, peas, green beans, tomatoes and strawberry) and fruits (dates, orange and lemon) were selected for this study as the most popular crops for Egyptians.

The situation of contamination with pesticide residues and number of samples analyzed from each commodity, number of contaminated samples,

the minimum, maximum and mean values of each pesticide in each commodity are presented in Tables 1, 2 and 3 detected in the samples randomly collected from different commodities available in the markets using the multiresidue method S19. The residue concentration is referred to the maximum residue limits (MRLs) of the Codex Committee on Pesticide Residues (CODEX MRLs) which is adopted and applied in Egypt by the Egyptian Standardization Organization for the safety of Egyptian consumer. The results indicate that only one green beans sample (0.613 mg/kg) violated the MRL of carbendazim (0.5 mg/kg) in green beans. Considering international trade and referring to the MRLs of the EU which is the major importer from Egypt and where the EU MRLs are the most restricted worldwide. The data in Tables 1, 2 and 3 revealed violation in 12 samples, i.e., profenefos in caraway, malathion in dill, ethion and propargite in strawberry, carbendazim in green beans, methomyl in pepper, profenofos in lemon and quinalphos in oranges.

Table 4 demonstrates the miss use of pesticides where most of the detected pesticide residues are either not registered for use on the crop contaminated with it or not registered for use at all in the country. Figure (1a) illustrates the situation of samples contamination referring to the total number of samples analyzed. Sixty (45.45%) of the samples were free from contamination with pesticide residues analyzed, seventy two (54.55%) were contaminated, from which 79.17% contaminated with residues from one pesticide, 11.11% with 2 residues and 9.72% with more than 2 residues.

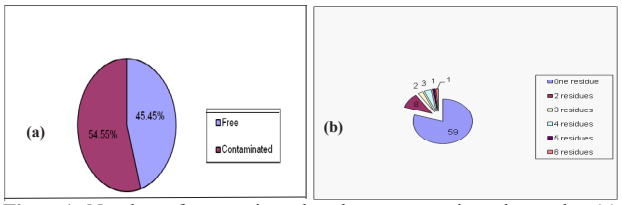

Figure1. Number of contaminated and non-contaminated samples (a) and number of samples contaminated with 1, 2, 3 or more pesticide residues (b)

Figure (1b) represents the number of samples contaminated with 1, 2, 3 or more pesticides residue. Fifty seven out of 132 samples analysed contained residues from one pesticide, 8 samples contained residues from 2 pesticides and 7 samples contained 3 or more pesticide residues. The last category includes 2 caraway and one fennel samples contained 4 pesticide residues, one sample of marjoram contained 5 pesticide residues, one mint sample comprised 6 pesticide residues. The present data indicate that the percentage number of samples contained 2 pesticides from all samples analyzed was 6.06% and those contained three or more was 5.3% a situation that need to be carefully considered.

Discussion

Residues of 17 pesticides were detected in the analyzed samples, i.e., chlorpyrifos, diazinon, malathion, profenofos, sulfur, chlorpyrifos-methyl, carbendazim, DDE-p,p, cypermethrin, ethion, propargite, permethrin, L-cyhalothrin, methomyl, phenpropathrin, quinalphos and pirimiphos-methyl. The discussion of the present study was focused on two areas, i.e., quantitative evaluations of the pesticide residue results compared to the Maximum Residue Limits (MRLs) of the Codex Committee which is the standard that supposed to be followed locally to protect the Egyptian consumers. The pesticide residue results were also compared to the EU (2010) MRLs which apply on the Egyptian exports to the European countries.

The second area is the concern on the Good Agricultural Practice (GAP) of the pesticide registered for use in the country on each specific crop under investigation. The situation of human risk of the detected pesticides in light of the world concern on carcinogenicity, endocrine disruption and cumulative risk (Cocktail effect) is also discussed.

It could be concluded from the data in Table 4 that only two pesticides are registered and permitted for use on the target crop which are methomyl on pepper and malathion on orange. Malathion registered and recommended for use on orange only as killing bags not supposed to leave residues on orange fruits. Other pesticides of which their residues were detected on the different crops are not officially permitted for use on them. In case of aromatic and medicinal plants, no pesticides are recommended and permitted for application on such group of plants. Ethion, permethrin, propargite and quinalphos are not registered for use in Egypt. DDT is banned in the 1970's. Presence of residues of non registered pesticides is alarming the need to strengthen the regulations and control on the pesticides illegally entering the country.

Residues of registered pesticides detected on a crop where they are not permitted for use, it could be due to contamination from adjacent fields applied those pesticide or from soil or water contamination. Violation of MRLs observed in Tables 1, 2 and 3 might indicate deviation from Good Agriculture Practice (GAP) where the pre-harvest intervals (safety intervals) are not followed or the rate of application and the concentration are not adjusted to the recommendation of the GAP. The data indicate a pressing need to adopt the GAP and the official

Table 1. Levels of pesticide residues of some herbs collected from Egyptian local markets

Commodity	Total number of samples	Number of contaminated samples	Detected pesticide	Number of contaminated samples with each pesticide	Minimum (ppm)	Maximum (ppm)	Mean (ppm)	MRL (ppm)			
								Codex (2010)	Violation	EU (2010)	Violation
Fennel	2	2	Chlorpyrifos	1	0.072	0.072	0.072	5.00		5	0
			Diazinon	1	0.010	0.010	0.010	5.00		5	
			Malathion	1	0.043	0.043	0.043	2.00		2	
			Profenofos	1	0.057	0.057	0.057			0.1	
Anise	3	2	Malathion	2	0.012	0.042	0.027	2.00		2	
			Sulfer	1	8.563	8.563	8.563				
Basil	6	4	**Chlorpyrifos**	3	0.010	0.036	**0.023**	**1.00**		0.5	
			Sulfur	3	0.050	0.496	0.310				
Caraway	5	3	Malathion	2	0.014	0.029	0.022	2.00		2	
			Chlorpyrifos	3	0.014	0.050	0.027	5.00		5	
			Chlorpyrifos-me	1	0.016	0.016	0.016	1.00		1	
			Profenofos	2	0.015	0.270	0.143			0.1	1
			Sulfur	1	0.025	0.025	0.030				
Chamomile	1	1	Chlorpyrifos	1	0.010	0.010	0.010	1.00		0.5	
Marjoram	6	6	Chlorpyrifos	5	0.015	0.147	0.050	1.00		0.5	
			Diazinon	1	0.010	0.010	0.010	0.10		0.02	
			Malathion	2	0.021	0.043	0.032	1.00		0.5	
			Profenofos	2	0.011	0.011	0.019			0.1	
			Sulfur	1	0.096	0.096	0.096				
			Carbendazim	1	0.039	0.039	0.039			0.1	
Dill	7	6	Diazinon	1	0.010	0.010	0.010	0.10		0.02	
			Malathion	2	0.010	0.530	0.270	1.00		0.5	1
			Chlorpyrifos	2	0.015	0.017	0.016	1.00		0.5	
			DDE-p,p'	1	0.018	0.018	0.018			0.5	
Mint	9	9	Chlorpyrifos	9	0.011	0.063	0.029	1.00		0.5	
			Chlorpyrifos-methyl	1	0.016	0.016	0.016	0.30		0.1	
			Diazinon	1	0.010	0.010	0.010	0.10		0.02	
			Malathion	3	0.013	0.043	0.031	1.00		0.5	
			Profenofos	1	0.057	0.057	0.057			0.1	
			Cypermethrin	1	0.050	0.050	0.050	0.10		0.1	
			Sulfur	3	0.096	12.071	4.948				
Hibisicus	1	0	ND	ND	ND	ND	ND				
Total	40	33									2

* MRLs refer to maximum residue limits issued by European Union Pesticides Database(2010).
** MRLs refer to maximum residue limits issued by Codex (2010).
ND means non-detected pesticide residue.
Expanded Uncertainty measurement ± 50%.

Table 2. Levels of pesticide residues of some vegetables collected from Egyptian local markets

Commodity	Total number of samples	Number of contaminated samples	Detected pesticide	Number of contaminated samples with each pesticide	Minimum (ppm)	Maximum (ppm)	Mean (ppm)	MRL (ppm)			
								Codex (2010)	Violation	EU (2010)	Violation
Strawberry	12	8	Ethion	3	0.015	0.047	0.034			0	3
			Propargite	1	0.023	0.023	0.023			0	1
			Permethrin	2	0.021	0.045	0.033	1.00		0.1	
			Profenofos	2	0.010	0.037	0.024			0.1	
			Chlorpyrifos	1	0.050	0.050	0.050	0.30		0.2	
Tomato	5	0	ND	0							
Onion	6	2	Sulfur	1	1.717	1.717	1.717				
			L-Cyhalothrin	1	0.073	0.073	0.073	0.20		0.2	
Cucumber	7	0	ND	0							
Lettuce	2	0	ND	0							
Okra	2	0	ND	0							
Peas	2	0	ND	0							
Green beans	7	1	Carbendazim	1	0.613	0.613	0.613	0.50	1	0.2	1
Pepper	11	4	Sulfur	2	0.091	0.165	0.128				
			Methomyl	2	0.323	0.348	0.336	0.70		0.1	2
Total	**54**	**15**							1		**7**

* MRLs refer to maximum residue limits issued by European Union Pesticides Database(2010).
** MRLs refer to maximum residue limits issued by Codex (2010).
ND means non-detected pesticide residue.
Expanded Uncertainty measurement ± 50%.

Table 3. Levels of pesticide residues of some fruits collected from Egyptian local markets

Commodity	Total number of samples	Number of contaminated samples	Detected pesticide	Number of contaminated samples with each pesticide	Minimum (ppm)	Maximum (ppm)	Mean (ppm)	MRL (ppm)			
								Codex (2010)	Violation	EU (2010)	Violation
Dates	3	0	ND	0							
Lemon	4	2	Profenofos	2	0.156	0.165	0.161			0.1	2
Orange	31	22	Fenpropathrin	1	0.045	0.045	0.045			2	
			Chlorpyrifos	5	0.010	0.072	0.040	1.00		0.3	
			Malathion	4	0.012	0.036	0.028	7.00		7	
			Quinalphos	1	0.109	0.109	0.109			0.1	1
			Pirimiphos-Me	8	0.026	0.167	0.072			1	
			L-Cyhalothrin	2	0.016	0.021	0.019			0.1	
			Cypermethrin	1	0.014	0.014	0.014	2.00		2	
			Ethion	1	0.017	0.017	0.017			2	
Total	38	24									3

* MRLs refer to maximum residue limits issued by European Union Pesticides Database(2010).
** MRLs refer to maximum residue limits issued by Codex (2010).
ND means non-detected pesticide residue.
Expanded Uncertainty measurement ± 50%.

Table 4. Pesticide /commodity combination detected in analyzed samples and situation of registration

Pesticide (common name)	Commodity	Registration status
Carbendazim	Marjoram	x
	Green beans	x
Chlorpyrifos	Fennel	√
	Basil	√
	Caraway	√
	Chamomile	√
	Marjoram	√
	Dill	√
	Mint	√
	Strawberry	√
	Orange	√
Chlorpyrifos-methyl	Caraway	√
	Mint	√
Cypermethrin	Orange	√
	Mint	√
Diazinon	Fennel	√
	Marjoram	√
	Dill	√
	Mint	√
Ethion	Strawberry	x
	Orange	x
Fenpropathrin	Orange	√
Lambda-Cyhalothrin	Orange	√
	Onion	√
Malathion	Fennel	√
	Anise	√
	Caraway	√
	Marjoram	√
	Dill	√
	Mint	√
	Orange	Recommended as killing bags only
Methomyl	Pepper	Recommended
Permethrin	Strawberry	x
Pirimiphos-Methyl	Orange	√
Profenofos	Fennel	√
	Caraway	√
	Marjoram	√
	Mint	√
	Strawberry	√
	Lemon	√
Propargite	Strawberry	x
Quinalphos	Orange	x
DDT p,p	Dill	x

X= not registered in Egypt.
√= registered for use on other crops but not recommended for use on that particular crop.

recommendation of the authorized pesticides. This could be achieved through educating and licensing farmers and applicators especially for the application of highly risk pesticides.

The amounts of pesticides remained as residues in food are miniscule sometimes only 1 millionth of a kilogram. The official safety limits allowed in our daily intake are also incredibly small. Pro-pesticide groups say this is equivalent to a fraction of a teaspoonful in an Olympic swimming pool and we shouldn't worry about such very low levels. This is misleading because many of the chemical messengers in our bodies function at precisely these minute quantities, of ppm or even ppb. Scientists cannot say for sure that there is ever a "safe" level of pesticide residues in food (Holland *et al*., 2008).

Cumulative risk assessment (cocktail effect)

The cocktail effect means that the current process by which governments decide on safe levels, i.e., via a 'risk assessment', where single chemicals are considered separately, ignores the reality that people and wildlife are constantly exposed to many chemicals simultaneously. This process significantly underestimates the risk to human health from the real-life cocktail exposure. Scientists are therefore now urging public authorities to assess the combined risks of chemicals together.

Pesticide residue data are very useful in the studies concerned with cumulative risk assessment. Governments of developed countries used to publish data to determine what foods have been tested and what pesticides were found. They look at how often pesticides at any level were detected, how frequently legal levels were exceeded and how often more than one pesticide was found on a sample (AAAS Risk Policy, 2003; Payne-sturges *et al*., 2009)

The pesticide residues detected in the present study (17 chemicals) belong to different chemical groups. Compounds that are sharing a common mechanism of action are those belonging to the organophosphate and the carbamate groups. Both groups are acetyl-cholinestrase inhibitors. Chlorpyrifos, diazinon, malathion, profinofos, chlorpyrifos-methyl, ethion, phenpropathrin, quinalphos and pirimiphos-methyl are organophosphorus pesticides and propargite and methomyl are from the carbamate group.

The development in the risk assessment of cumulative action and methods of its calculations should be closely followed up in order to apply it (when available with low uncertainty) in the cases of detection of more than one pesticide in the samples analyzed. Such assessment should follow a case by case study.

Endocrine disruption

Some pesticides are suspected of being endocrine (hormone) disruptors. These chemicals affect parts of the body's hormone systems and can lead to an increase in birth defects, sexual abnormalities and reproductive failure, and may increase the risk of cancers of reproductive organs (Hayes *et al*., 2006). There is increasing evidence both from epidemiology studies and animal models that specific endocrine-disrupting compounds may influence the development or progression of prostate cancer (Raghow *et al*., 2002). In large part, these effects appear to be linked to interference with estrogen signaling, either through interacting with Estrogen Receptors (ERs) or by influencing steroid metabolism and altering estrogen levels within the body (Steiner and Pound, 2003). In humans, epidemiologic evidence links specific pesticides, PCBs and inorganic arsenic exposures to elevated prostate cancer risk (Prins, 2008).

Referring to the List of Lists (2009), four of the pesticides detected in cereal and cereal products are listed at different stages as endocrine disruptors. According to this list quinalphos in consideration as disruption as grade one where at least one study providing evidence of endocrine disruption in an intact organism. While, malathion, carbendazim, diazinon and permethrin have potentials for endocrine disruption as grade two.

However, malation is presented in 18 out of 132 samples analyzed, carbendazim in 2 samples, diazinon in 4 samples and permethrin in 2 samples. Quinalphos was only detected in one sample. It is of utmost importance to consider the risk to humans and children that might result from the dietary intake of endocrine disruption pesticides with first priority given to malathion as the major endocrine disruptor detected in the food samples analyzed. However, the registration and use of endocrine disruptors like malathion, diazinon, carbendazim, permethrin and quinalphos should be reconsidered by risk managers for the sake of consumer safety and reduce health hazards.

Potential human carcinogen

Another concern about the long-term effects of certain pesticides in food is cancer. The link between pesticide intake and cancer cannot definitely be proved, however, the authorities make decisions on whether to license a particular pesticide or not, taking into consideration the results from animal laboratories if might cause cancer. Study of pesticide residue levels suggested that children under five years could rapidly build up their cancer risk from residues in

food in the first few years of life as their food intake is very different from that of adults (Boobis *et al*., 2008).

According to the data in Tables 1, 2 and 3, it could be concluded that pesticides classified under EPA category (2) permethrin, likely to be carcinogenic to humans and (3) malathion, suggestive evidence of carcinogenicity but not sufficient to assess human carcinogenic potential, (B2) DDT and propargite, indicates sufficient evidence in animals and inadequate or no evidence in humans, (C) carbendazim Possible Human Carcinogen. EU classification set DDT and propargite under category (3) possible risk of irreversible effects (cancer). The International Agency for Research on Cancer (IARC) classified malathion and permethin under group (3) not classifiable as to carcinogenicity in humans and DDT under group (2B) possibly carcinogenic to humans (IARC, 1991).

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