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Pesticide residues in some Egyptian spices and medicinal plants as affected by processing

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Abstract

Pesticide residues were determined in Egyptian spices and medicinal plants. For this purpose, a total of 303 samples, which represent 20 different plants were collected from sources in Egypt and several shipments All the collected samples were analyzed for the determination of organophosphorus and organochlorine residues. The obtained results showed the predominance of malathion in most of the analyzed samples. The detected concentrations of it in jews mallow, dill, celery, tea, caraway, chamomile and saffron exceeded the maximum permissible levels (MPLs), as did the concentrations of dimethoate in caraway and chamomile samples. Low levels of profenofos, pirmiphos-methyl, chloropyrifos. parathion and diazinon were determined in the analyzed samples. Residues of lindane, aldrin, dieldrin, DDT, chlordane and endrin in chamomile samples exceeded the MPLs. Residues of aldrin and dieldrin in karkade were higher than the MPLs, as was chlordane in peppermint. Residues were not detected in the watery extract when the medicinal plant was boiled in water. Also, immersing the plants in hot water transferred some pesticide residues to the aqueous extract.

1. Introduction

Spices and medicinal plants are widely used as raw materials for pharmaceutical preparations (Galenic products) and as a supplement for dietetic products and especially for ''self medications'' in the general population. These plants are susceptible to insect and disease attacks, so pesticides are widely used for protection. Therefore, residues of pesticides could reach and affect consumers especially when they are freshly consumed.

The published research on pesticide residues in crude herbal materials indicate that the presence of chlorinated pesticide residues is quite common. DDT and its derivatives, γ-HCH and other HCH isomers, HCB, and cyclodiene derivatives such as aldrin, dieldrin, heptachlor and its epoxide were reported to occur in these plants (Ali, 1983; Benecke, Brotka, Wijsbeck, Frank, Bruins & Dezeeuw, 1986, 1989; Kwon, Dicke & Thier, 1986; Pluta, 1988; Schilcher, 1985). Other potentially contaminating pesticides include organophosphates, carbamate Insecticides and herbicides, dithiocarbamate fungicides and triazine herbicides (Beneck, Ennet & Frauenberger, 1987; Ennet, 1989; Schilcher, 1985; Schilcher, Peters & Wank, 1987). In addition, polychlorinated biphenyls have been reported to occur in raw herbal materials as a result of general environmental pollution (Beneck et al., 1988).

Many studies have been carried out in different countries on spice and medicinal plants. In Germany, it was reported that the residues of DDT and its derivatives, HCH, dimethoate, and parathion methyl predominated as contaminants (Ali, 1983; Beneck & Ortwein, 1992; Beneck at al., 1987; Schilcher, 1982). On the other hand, DDT, HCH, aldrin, dieldrin, and heptachlor were detected in different samples collected from India (Kannan, Tanobe, Ranesh, Subramanian & Tatsukawa, 1992; Kaphalia, Takroo, Mehrotra, Nigam & Seth, 1990). Other studies in developed countries indicated that the organochlorine pesticide residues were the main compounds detected in spice and medicinal plants (Sullivan, 1980). The results of Dogheim, Almaz, Takia and Youssef (1986) showed the predominance of HCH residues in the samples collected from shipments for exportation from Egypt. The same study also found the prevalence of DDT, but its levels were below the maximum permissible limits. To control pesticide residues in

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food, many countries restricted the use of organochlorine pesticides, mainly in food applications, because of their long persistence.

In consequence of convenient climatic factors in Egypt, cultivated herbs are of high quality (concerning the active substances) and this encourages exportation. In recent years the cultivation of medicinal and aromatic plants has been pursued with increasing interest. In Egypt, up till now, studies dealing with the detection of pesticide residues in spice and medicinal plants have been lacking.

This study was conducted to reveal and draw attention to the great problem of environmental pollution, in particular by pesticide residues in spice and medicinal plants from shipments for exportation from Egypt, to ensure safety and quality. In addition, the study aims to shed light upon the behaviour of pesticide residues in medicinal plants during the extraction by hot and boiling water.

2. Materials and methods

2.1. Materials

2.1.1. Sample collection

A total of 303 samples, which represented 20 different types of spices and medicinal plants, were collected from sources in Egypt, many with different growing seasons and each with its own agricultural practices and several shipments. Eleven types belong to the leafy group, six to the fruity group and three belong to the flowery group. The collected samples were kept in aluminium foil and frozen until analysis. The various collected samples (item, scientific name, number) are presented in Table 1.

Table 1
Spice and medicinal plant samples collected from several selected shipments in Egypt

Item	No. of collected samples	Scientific name
Leaves		
Geranium	20	*Pelargonium graveolens* L.
Basil	20	*Ocimum basilicum* L.
Marjoram	20	*Marjorana hortensis* L.
Peppermint	20	*Mentha piperita* L.
Spearmint	20	*Mentha viridis*
Jews mallow	10	*Corchorus oritorius*
Dill	10	*Anethum grveolens* L.
Celery	10	*Apium graveolens* Mill.
Parsley	10	*Petroselinum sativum* Hoffm
Cumin	10	*Cuminum cyminum* L.
Tea	6	*Thea sinensis* Linn.
Fruits		
Caraway	15	*Carum carvi* L.
Anise	15	*Pimpinella anisum* L.
Fennel	15	*Foeniculum vulgare* L
Coriander	15	*Coriander sativum* L
Dill	5	*Anethum graveolens* L.
Black pepper	5	*Piper nigrum*
Flowers		
Chamomile	66	*Matricaria chamomila* L.
Karkade	6	Rosella jamica
Saffron	5	*Crocus sativus* Linn

2.1.2. Standards

Standards of malathion, dimethoate, profenofos, pirimiphos-methyl, chlorpyrifos, parathion, diazinon, lindane, aldrin, dieidrin, heptachlor, heptachlor epoxide, chlordane, HCB, endrin, endosulfane, 1,1,1-trichloro-2-(2-chlorophenyl)-2-(4-chlorophenyl)ethane(o.p′-DDT), 1,1, 1-trichloro-2, 2-bis (4-chlorophenyl) ethane (p,p′-DDT), 1, 1-dichloro-2,2-bis(4-chlorophenyl)ethane (o,p′-DDD), 1,1-dichloro-2,2-bis-(p-chlorophenyl) ethane (p,p′-DDD), 1,1-dichloro-2-(2-chlorophenyl-2-(4-chioro-phenyl) ethane (o,p′-DDE) and 1,1-dichloro-2,2-bis (4-chloro phenyl) ethane (p,p′-ODE) were purchased from Chem. Service, Inc. (West Chester, PA).

2.2. Methods

2.2.1. Preparation of the plant watery extract

Medicinal plants; i.e. spearmint, caraway, anise, chamomile, and karkade were extracted in hot water by two different methods as commonly used at home. In the first method, 2 g of the dry plant were left to boil in 100 ml deionized water for 5 min in a glass beaker. In the second method, 2 g of the dry sample was immersed in 100 ml of hot deionized water for 5 min (tea method). The liquid extracts in the two previous procedures were filtered separately and were prepared for analysis.

2.2.2. Determination of pesticide residues in the plants

Residues of pesticides in spice and medicinal plant samples were determined according to the method of WHO/Pharm. (1992) as follows: The plant material was ground and mixed thoroughly. A weight of 20–50g of the ground sample was blended with 350 ml of acetonitrile/water (65:35) for 5 min at a high speed. The extract was combined, filtered and measured; then it was shaken vigorously in a separating funnel with 100 ml of light petroleum ether for 1–2 min to transfer pesticide residues to the light petroleum ether layer. The extract of petroleum ether was transferred directly to a column of activated florisil (60/100 PR) leaving it to pass through the column at a rate of 5 ml/min, then it was eluted with a mixture of diethyl ether/light petroleum ether. The final eluate was dried to a definite volume for gas chromatographic analysis. Analysis was carried out with Gas Chromatography (Hewlett-Packard, Model 5890) equipped with a ^{63}Ni Electron Capture Detector (ECD) and a Flame Ionization Detector (FID). An HP-I (methyl silicon fluid) capillary column (30 m×0.25 mm, 0.25-μm film thickness) was used. The chromatograms were integrated with an HP-3392A integrator.

The applied temperature programming was as follows: initial temperature, 80°C; hold for 2 min; increased to 160°C at 3°C min^{-1}; hold 2 min; increase to 220°C at 5°C min^{-1}; hold 20 min Injection temperature 220°C, and detector temperature, 300°C. Percent recoveries were determined for this method, and all the results were corrected according to the recovery percent for each pesticide, separately.

2.2.3. Determination of pesticide residues in the watery extract

Pesticide residues were determined in the watery extract of the medicinal plants according to the AOAC (1995) method.

3. Results and discussion

3.1. Pesticide residues in spices and medicinal plants

Organophosphorus pesticides are widely used in agriculture and animal production for the control of various insects. These compounds have higher acute toxicity than chlorinated pesticides and they have the advantage of being more rapidly degraded in the environment. Organochlorine pesticides, which 20 years ago were being used in Egypt, are highly persistent. Most of them have been banned, yet their residues still appear as pollutants in food as well as in the environment. Residue levels of these compounds in spice and medicinal plants are listed in Tables 2–4.

The identities of organochlorine pesticide residues found in the leaf group samples of medicinal plants are given in Table 2. Examination of the data show that, in some analyzed samples, residues of some of the tested pesticides were below the detection limits. The other samples contained variously different levels of the residues. Residues of chlorpyrifos, parathion and diazinon were below the detection limits in all of the analyzed samples.

Among various organophosphorus pesticides in the present study. malathion is the predominant compound in most leaf group samples (Table 2). The detected levels of it varied greatly. For instance, the minimum value for it was detected in basil (0.093 mg/kg), and the maximum of 1.719 mg/kg was found in dill. Corresponding values for total DDT were 0.005 mg/kg in basil and 0.735 mg/kg in tea, respectively.

With respect to the fruit plant samples, Table 3 shows that residues of pirmiphos methyl, chloropyrifos. parathion and diazinon (as organophosphorus pesticides, OP), in addition to lindane, aldrin, dieldrin, heptachlor, heptachlor epoxide, chlordane, HCB, endrin and endosulfane (as organochlorine pesticides, OC), were below the detection limits in all the analyzed samples. In contrast, malathion (OP) predominated in all the analyzed samples, followed by dimethoate and profenofos. The minimum detected residue of malathion was detected in fennel (0.074 mg/kg), and the maximum value was found in dill (0.686 mg/kg).

Despite this trend, a maximum of 1.756 mg/kg (caraway), and 0.461 mg/kg (dill) and a minimum of 0.306 mg/kg (black pepper) and 0.061 mg/kg (fennel) were recorded for dimethoate and profenofos, respectively. Derivatives of DDT were detected in caraway, anise, fennel and coriander to be 0.009, 0.217, 0.204 and 0.071 mg/kg, respectively. It was not detected in dill and black pepper samples.

In regard to the flower group samples, Table 4 indicates that malathion is the predominant compound. The highest mean levels were detected in chamomile followed by saffron and karkade. which recorded 2.187, 0.669, and 0.364 mg/kg, respectively. However, dimethoate and profenofos were detected in chamomile and karkade at levels of 1.779 and 0.362 and 0.008 and 0.818 mg/kg, respectively. But pirmiphos methyl was detected only in chamomile samples which showed mean value 0.105 mg/kg. The other organophosphorus pesticides under investigation were not detectable in the flower group samples. The results obtained (in the same table) indicated that various samples contained organochlorine pesticides at different levels. Chamomile samples showed the highest mean levels of these compounds. However, Saffron contained aldrin (0.066 mg/kg) and karkade contained lindane, aldrin, and dieldrin at levels of 0.068. 0.089, and 0.110 mg/kg, respectively.

The data show the decreased concentrations of the residues of DDT and its derivatives, which were detected in all the samples of spice and medicinal plants under investigation except chamomile, with low frequencies. DDT is one of the most important and probably the most common insecticide in the world, as well as in Egypt, where it has been extensively used for insect control in agriculture. Commercial products of 50 or 70% were usually used, which contain mainly p.p′-DDT, while o.p′-DDT is present as the major contaminant of the product. The decreased levels of DDT and its derivatives in the current study can be due to the Agricultural Ministry of the Egypt Government who advised farmers not to use DDT. However, significantly elevated levels of this insecticide were detected in samples under investigation. Probably this insecticide is still being applied, despite the Ministry's recommendation. Continuous contamination with DDT might be due to its presence at low concentrations in the irrigation water coming mainly from the River Nile (El-Mekkawi, 1994) which runs through other developing countries where organochlorines are still in use. Aldrin and dieldrin were expected to appear in some types of plants. This may be due to their long use in Egypt for agriculture. Both are insecticides, and it is well known that, aldrin converts to

Table 2
The distribution levels (mg/kg±S.D.) of pesticide residues detected in Egyptian spice and medicinal plant samples (Leave group)

Pesticide types	Geranium	Basil	Marjoram	Peppermint	Spearmint	Jews mallow	Dill	Celery	Parsley	Cumin	Tea
Organophosphorus											
Malathion	0.146±0.122	0.093±0.089	–	0.422±0.24	0.368±0.168	0.516±0.490	1.719±0.684	0.640±0.221	–	0.246±0.116	0.608±0.480
Dimethoate	0.618±0.441	0.712±0.360	–	0.351±0.112	0.516±0.411	0.849±0.349	–	–	–	0.104±0.074	0.266±0.116
Profenofos	–[a]	0.159±0.124	–	0.40±0.211	0.681±0.311	0.246±0.206	0.279±0.116	0.101±0.094	0.009±0.006	0.116±0.112	–
Pirmiphos-Me	–	–	–	0.122±0.116	–	–	0.062±0.011	0.040±0.024	0.112±0.101	–	–
Chlorpyrifos	–	–	–	–	–	–	–	–	–	–	–
Parathion	–	–	–	–	–	–	–	–	–	–	–
Diazinon	–	–	–	–	–	–	–	–	–	–	–
Organochlorines											
Lindane	0.051±0.022	–	–	0.128±0.116	0.106±0.100	–	–	–	–	–	–
Aldrin	–	–	–	0.053±0.033	–	–	–	–	–	–	–
Dieldrin	0.036±0.022	0.027±0.016	0.014±0.012	0.015±0.006	0.018±0.011	0.026±0.011	–	–	–	–	–
Heptachlor	–	0.013±0.012	0.045±0.038	0.124±0.118	–	–	–	–	–	–	–
Hep. Epoxide	0.026±0.020	0.015±0.008	0.023±0.020	–	0.028±0.016	–	0.022±0.016	0.01 1±0.008	–	–	0.068±0.046
Chlordane	–	–	–	0.197±0.168	–	–	–	–	–	–	–
HCB	–	0.153±0.010	0.256±0.212	0.139±0.110	–	–	–	–	–	–	–
Endrin	–	0.018±0.012	0.016±0.014	0.083±0.074	–	–	–	–	–	–	–
Endosulfane	–	–	–	–	–	–	–	–	–	–	–
o.p′-DDT	0.166±0.122	–	0.368±0.112	0.254±0.116	–	0.036±0.022	–	–	0.116±0.082	0.094±0.068	0.147±0.156
p.p′-DDT	0.206±0.200	–	–	–	–	0.098±0.061	–	–	0.024±0.020	–	0.220±0.200
o.p′-DDD	–	–	–	0.108±0.081	–	–	–	–	–	–	–
p.p′-DDD	–	–	–	0.084±0.07	–	–	–	–	–	–	–
o.p′-DDE	0.096±0.041	–	0.10±0.038	0.063±0.051	0.168±0.118	0.184±0.180	0.027±0.011	0.011±0.006	0.061±0.047	–	0. 160±0.110
p.p′-DDE	0.146±0.106	0.005±0.003	0.005±0.003	0.024±0.016	0.246±0.169	0.286±0.221	0.027±0.016	0.072±0.050	–	–	0.208±0.166
Total DDT	0.614	0.005	0.473	0.529	0.414	0.604	0.054	0.083	0.201	0.094	0.735

[a] –, below the detection limits.

Table 3
The detected levels (mg/kg ± S.D.) of pesticide residues in Egyptian spice and medicinal plant samples (fruit group)

Pesticide types	Caraway	Anise	Fennel	Coriander	Dill	Blackpepper
Organophosphorus						
Malathion	0.625±0.242	0.306±0.118	0.074±0.056	0.318±0.221	0.686±0.241	0.480±0.311
Dimethoate	1.756±0.689	0.408±0.206	0.614±0.406	–	–	0.306±0.112
Profenofos	0.284±0.108	–	0.061±0.008	0.121±0.094	0.461±0.230	–
Pirmiphos-Me	–[a]	–	–	–	–	–
Chlorpyrifos	–	–	–	–	–	–
Parathion	–	–	–	–	–	–
Diazinon	–	–	–	–	–	–
Organochlorines						
Lindane	–	–	–	–	–	–
Aldrin	–	–	–	–	–	–
Dieldrin	–	–	–	–	–	–
Heptachlor	–	–	–	–	–	–
Hep. Epoxide	–	–	–	–	–	–
Chlordane	–	–	–	–	–	–
HCB	–	–	–	–	–	–
Endrin	–	–	–	–	–	–
Endosulfane	–	–	–	–	–	–
o.p.′-DDT	–	–	0.091±0.066	–	–	–
p.p.′-DDT	–	–	0.102±0.061	–	–	–
o.p.′-DDD	–	–	0.011±0.007	–	–	–
p.p.′-DDD	–	–	–	0.011±0.006	–	–
o.p.′-DDE	0.009±0.006	0.116±0.081	–	0.024±0.009	–	–
p.p.′-DDE	–	0.101±0.066	–	0.036±0.026	–	–
Total DDT	0.009	0.217	0.204	0.071	–	–

[a] –, below the detection limits.

dieldrin by an epoxidation process in biological systems (Rumsey & Bond, 1974) and, therefore, dieldrin is expected to be found in relatively higher levels than aldrin. The reason for the contamination of the samples by heptachlor and its epoxide, in spite of the complete bay, might be described to the transformation of chlordane which is another organochlorine pesticide restricted in use to control wood borers in Egypt (EI-Mekkawi).

Similar results were obtained by Pluta (1988) who found that samples collected from Poland within the periods of 1980–1988 contained DDT and its derivatives at levels ranging from 0.2 to 3.2 ppm. In India, further results were obtained by Kannan et al. (1992). They found DDT, aldrin, dieldrin, and heptachlor at mean levels (ng/g) 62.0, 0.22, 2.1, 20.0, and 0.46, respectively in five types of spices (fennel, coriander, cumin, pepper, and chilies). In Germany, Ali (1983, 1987), Beneck and Ortwein (1992), Schilcher et al. (1987) and Zambo, Tetenyi and Bernath (1989) found DDT and HCH in different commercial vegetable drugs. Acceptable limits of the residues, established at 0.5 mg/kg by the German Pharmacopia, were exceeded in some samples. In Egypt, Dogheim et al. (1986) indicated that basil, marjoram, coriander, mentha Spp., and anise samples, collected from shipments for exportation, showed HCH levels of 0.53, 0.23, 0.26, 0.27 and 0.36 mg/kg, respectively. DDT was the next in prevalence, but in most cases. its level was below the maximum permissible level of 1.0 mg/kg. Another result, by Kaphalia et al. (1990) and Robin, Guillet, Ferry and Collombel (1978), reported that the samples collected from different countries contained residues of organochlorine pesticides. In Poland, Lutomski and Debska (1974) reported levels of DDT up to 0.47 ppm in eight spices and herbs, including allspice, basil, and black pepper Illes, Mestres, Tourte, Campo and Illes (1976) reported similar levels of the chlorinated hydrocarbons in several herbs.

From this work it can be seen that malathion, is detected in jews mallow, dill, celery, tea, caraway, chamomile and saffron at levels higher than the maximum permissible levels (MPLs) recorded by the Egyptian Organization for Standardization and Quality Control (E.O.S., 1991) and Pharmeuropa (1993). Also, dimethoate was higher in caraway and chamomile samples than the MPLs for E.O.S. It can also be seen that lindane, aldrin and dieldrin, DDT, chlordane and endrin were detected in chamomile samples at levels higher than the MPLs recorded by E.O.S. Also, aldrin and dieldrin in karkade as well as chlordane in peppermint were detected at levels higher than the MPLs recorded by E.O.S.

Table 4
The detected levels (mg/kg±S.D.) of pesticide residues in Egyptian spice and medicinal plant samples (flower group)

Pesticide types	Chamomile	Karkade	Saffron
Organophosphorus			
Malathion	2.19±0.986	0.364±0.206	0.669±0.489
Dimethoate	1.78±0.648	0.008±0.006	–
Profenofos	0.362±0.344	0.618±0.480	–
Pirmiphos-Methyl	0.105±0.088	–	–
Chlorpyrifos	–[a]	–	–
Parathion	–	–	–
Diazinon	–	–	–
Organochlorines			
Lindane	0.801 ± 0.710	0.068±0.041	–
Aldrin	0.167±0.118	0.089±0.064	0.066±0.044
Dieldrin	0.167±0.042	0.110±0.009	–
Heptachlor	0.142±0.046	–	–
Hep. Epoxide	0.047±0.019	–	–
Chlordane	0.592±0.162	–	–
HCB	1.237±0.571	–	–
Endrin	0.142±0.116	–	–
Endosulfane	–	–	–
o.p′-DDT	0.139±0.097	–	–
p.p′-DDT	0.088±0.046	–	–
o.p′-DDD	0.305±0.169	–	–
p.p′-DDD	0.193±0.097	–	–
o.p′-DDE	0.262±0.176	–	–
p.p′-DDE	0.314±0.304	0.114±0.019	–
Total DDT	1.30	0.114	–

[a] –, below the detection limits.

These results, indicate that pesticides should be applied correctly using only the required amounts and following label directions.

3.2. Behaviour of pesticide residues in the medicinal plants during processing

Table 5 shows that, immersing the medicinal plants in hot water transfers pesticide residues to the watery extract in different ratios. For example, the extracted concentrations of profenofos in the watery extract of (caraway), profenofos (in anise), lindane (in chamomile) and endrin (in karkade) were 2.4, 9.9, 37.1 and 27.6%, respectively, of their initial concentrations in the plant. On the other hand, the other tested pesticides were completely absent in the aqueous extract. All the residues of investigated pesticides in the watery extract were below the detection limits after boiling the medicinal plant in water.

From these results, it could be concluded that the behaviour of pesticide residues during preparation in water depends on the type of herbs, the type of pesticide and on the applied treatment. The disappearance of pesticide residue from boiling extract could be due to decomposition by the effect of heat, the stronger adsorption of pesticide onto plant tissues and/or the poor solubility of pesticides in water (Ali, 1983). The level of contamination is not only reduced by extraction with boiling water, but also by ethanolic extraction (Ennet, 1989; Schilcher, 1982).

Table 5
Behaviour of pesticide residues in medicinal plants during processing

Medicinal plant	(1)[a]	(2)[b]	(3)[c]
Spearmint			
Lindane	0.063	nd	nd[d]
Profenofos	3.46	nd	nd
Caraway			
Profenofos	3.49	3.411	Nd
Anise			
O,p′-DDE	0.081	nd	nd
p,p′-DDT	0.355	nd	nd
Pirmiphos Methyl	0.789	nd	nd
Profenofos	0.893	0.812	nd
Chamomile			
Lindane	0.189	0.119	nd
o,p′-DDT	0.810	nd	nd
Karkade			
Endrin	0.930	0.673	nd

[a] Initial concentration before treatment (μg/2 g dry plant).
[b] Concentration in hot water (μg/100 ml watery extract).
[c] Concentration in boiling water (μg/100 ml watery extract).
[d] Below the detection levels.

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