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Investigation of Pesticides Residues in Some Medicinal Plants Collected from Local Markets in Jeddah, Saudi Arabia

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Abstract: Pesticides residues were estimated in some commonly used medicinal plants collected from different markets in Jeddah, Saudi Arabia ,these are; (Rosemary; Rosmarinus officinals L. & Sage; Salvia officinals L.), family Lamiaceae, (Anise; Pimpinella anisum L., Caraway, Carum carvi L. & Cumin; Cuminum cyminum L.) family Apiaceae, (Cinnamon; Cinnamomum verum L.) family Lauraceae, (Ginger; Zingiber officinale Roscoe.) family Zingiberaceae and (Tea ; Camellia sinensis L.) family Theaceae. It was found thatMalathion,Pirimiphos-methyl and profenofos predominated in most all investigated samples while fungicides were detected only in Cumin and Caraway samples in the form of azole compounds .(tebuconazole, propiconazole, flusilazole, difenoconazole) carbamate compounds (carbendazim) and other fungicides(iprodione, azoxystrobin, metalaxyl, flusilazole, thiophanate-methyl,ticyclazole, kresoxim-methyl and pendimethalin). Insecticides were dominated in Cumin, Caraway, Anise, Rosemary, Tea and Sage samples and mainly organophosphates (malathion, chlorpyrifos, profenofos, pirimiphos-methyl, ethion. The Ginger samples were free from pesticides while Cinnamon samples showed only the chloropyrifos<LOQ But in Anise difenoconazoleit was only in<LOQ. The medicinal plants and natural herbs must be used after the application of safety experiments and safety tests. It is recommended that everycountry use medicinal plants and spices (collected from natural habitats or cultivated) should have at least one control laboratory capable of performing the determination of pesticides residues and give a license for safety to be used in safe manner.

Keywords: Fungicides, Medicinal Plants, Insecticides, Jeddah, Safety, Saudi Arabia, Pollution

1. Introduction

A huge amounts of medicinal plants are consumed in our daily life, in pharmaceutical industry and as crude herbs where they seemed to be safe products by consumers eg; Curcuma, Cayenne pepper, Ginger, Anis, Mint, Onions, Fenugreek, and Cumin enhance the synthesis of bile acids in the liver and their excretion in bile, what beneficially accelerate the digestion and absorption of lipids. Most of medicinal plants enhance the pancreatic and digestive enzymes of gastric mucosa (Lipase, Amylase and Protease) [1]. Also the extracts from medicinal plants and spices accelerate the digestion and decrease the time of feed/food passage through the digestive tract [2&3]. Extracts of Curcuma, Red pepper, Black pepper, Cumin, cloves, Nutmeg, Cinnamon, Mint and Ginger showed anti -inflammatory effects in the rats [4]. Many bioactive compounds of medicinal plants and spices play a vital role in the prevention of Lipid peroxidation. Generally medicinal plants are seemed to be miraculously treat many diseases and never looked at as a possible source of toxic substances [5]. Residues of pesticide are found in all food items, vegetables, grains, fruits, eggs and fishes. Medicinal plants are liable to contain pesticides residues, which accumulate from Agriculture practice, treatment of soil during cultivation and administration of pesticides during storage of plants [6]. Insect

killer as DDT and its derivatives, g-HCH and other HCH isomers, HCB and cyclodiene derivatives such as aldrin, dieldrin, heptachlor and its epoxide were detected in crude herbal materials [7]. other potentially contaminating pesticides includes ; organophosphates, carbamate Insecticides and herbicides, dithiocarbamate fungicides and triazine herbicides were also detected in the daily used medicinal plants [8]. In addition polychlorinated biphenyls have been reported to occur in row herbal materials as a results of environmental pollution [9]. According to the use of pesticides become a routine work in growing these plants in many countries and the level of using of these chemical materials is not very well authorized in developing countries and may have bad impacts and toxic effects in human health if it used without precautions and safety legally percentages. Accordingly this study was planned to investigate the levels of different pesticides residues in most common used medicinal plants and herbs collected from Jeddah, Saudi Arabia, with special references to their impacts on human health.

2. Materials and Methods

Plant Samples were collected randomly from the Jeddah central market and immediately wrapped in aluminum foil, placed in an ice-chest kept at 4Ċ and sent to the laboratory for analysis .In the laboratory, similar samples were wholly bulked together and ground in a warring blender to obtain a homogenous composite. The used reagents are descried; A cetonitrile from Lab-scan HPLC, assay >99 (%, Methanol, 99.9 %HPLC grade Merck. Formic Acid, 98-100) % Riedel–de Haen, Ammonia solution, 33 % Riedel–de Haen, Sodium chloride, 99 % Riedel–deHaen, Disodium hydrogen citrate sesquihydrate, Fluka, Tri-sodium citrate di-hydrate, Fluka, Sodium chloride and anhydrous magnesium sulphate Merck, De-ionized water was produced by Milli-Q unit Millipore. Buffer-salt-mixture for second extraction and partitioning was prepared by weighing 4 ± 0.2 g of anhydrous magnesium sulphate , 1±0.05 g of sodium chloride, 1 ± 0.05 g of tri-sodium citrate dehydrate, 3.16 and 0.5±0.03 g of disodium hydrogen citrate, sesqui-hydrate into 25 ml glass tube. LC mobile phase was 10 mM ammonium format solution in methanol-water (1:9), pH 4±0.1. Sample dilution buffer was 10 mM ammonium format solution in methanol/water (1:1), pH 4 ± 0.1. Then the samples were prepared for pesticides analysis using a reference standard method for 387 pesticides with Purity of >95% [10]. The Pesticides extraction and clean-up were performed according to QuEChERS method as described by [11, 12 & 7]. where plant sample (10 g) was add in polyethylene (PFTE) 50 ml tube then 10 ml aceto-nitrile was add and shacked vigorously for one minute, then buffer-salt-mixture (4 g ± 0.2 g of magnesium sulfate anhydrous, 1 g ± 0.05 g of sodium chloride, 1 g ±0.05 g of tri-sodium citrate dehydrate and 0.5 g ± 0.03 g of disodium hydrogen citrate sesqui-hydrate) was added and shakes immediately for one minute. Samples were centrifuged at 4000 rpm for 5 minutes, supernatant (4 ml) of the clear solution was transferred to 50 ml round-bottomed flask and evaporated with rotary evaporator at 40 o C. Residues were re dissolved in 4ml (Methanol: ammonium format buffer 10 mM pH 4 (1:1). Samples were then filtered through0.45 µm syringe filter, the clear filtrate was used for injection into LC/MS/MS system.

Samples analysis for pesticides residues were performed using LC-MS/MS as follow, Agilent 7890 series gas chromatograph(Agilent technologist, Santa, Clara, CA) interfaced to an Agilent 5975 mass selective detector(MSD) Data acquisition, processing and instrumental control were performed by the Agilent MSD Chem. Station software (E.0200.493 version). A split/split less(S/SI) inlet was used with 1.8 mm id liner. Analyze samples were separated in an Aiglent HP-5MS capillary column(5%biphenyl/ 95% dimethyl siloxane), 30m, 0.25mm id, 0.25 µm film thicknesses. The inlet operating conditions were injection volume 1 µm, flow rate 1.3 ml/minute; the temperature program was set at79 Ċ for 0.25 minute, programmed to 300Ċ at 10 Ċ/minute, and kept at this temperature for 2 minute. The helium carrier gas flow was maintained at a constant of 17.296 psi The oven temperature program was 70 Ċ for 1 minute, programmed to 150 Ċat50 Ċ/minute then to 200 Ċ at 6 Ċ/minute. and finally to 280 Ċ at 16 Ċ/minute; it was kept at this temperature for 5 minute. Electron impact mass spectra in the full -scan mode were obtained at 70 eV; the monitoring was from m/z 50 to 400. The ion source and quadruple analyze temperature were fixed at 230 and 150 Ċ, respectively. The criteria of Quality Assurance Procedure of the Codex committee were followed as calibration curves for the two detectors were made at levels of 10,20,50,100 ,250 and 400 ug/kg-l Calibration curves were generated by plotting the relative responses of analyte (peak area of analyte/peak area of internal standard (IS)) to the relative concentration of analytes (concentration of analyze /concentration of IS) a constant amount internal standard, 5 µg kg-1was contained in the aliquot of the samples .the regression fit used for the calibration was the average response factor Recovery and reproducibility were evaluated by spiking pesticide standards to medicinal plants levels of 10, 50 &200 µg/kg where the analysis was performed in replicates of four at each levels and the average recovery varied between 81- 97. According to [13& 14]. The mean and standard deviation of results were represented in Tables (1&2).

Table (1). *Herbal plants under investigation; name, parts and medicinal properties.*

Herbal Plants	Part used	Medicinal properties
Caraway	Seeds	Caraway is used for digestive problems including heartburn, gas, loss of appetite, and mild spasms of the stomach and intestines. Caraway oil is also used to help people cough up phlegm, improve control of urination, kill bacteria in the body, and relieve constipation.
Cumin	Seeds	Cumin seeds have traditionally been noted to be of benefit to the digestive system, and scientific research is beginning to bear out cumin's age-old reputation.

Herbal Plants	Part used	Medicinal properties
Anise	Seeds	Anise it is greatly used in the form of lozenges and the seeds have also been used for smoking, to promote expectoration. The volatile oil, mixed with spirits of wine forms the liqueur Anisette, which has a beneficial action on the bronchial tubes, and for bronchitis.
Sage	Leaves, small stems and flowers	Sage is used for digestive problems, including loss of appetite, gas (flatulence),stomach pain (gastritis), diarrhea, bloating, and heartburn. It is also used for reducing overproduction of perspiration and saliva.
Rosemary	Leaves and flowers	The herb parts, especially flower tops contain phenolic anti-oxidant rosmarinic acid as well as numerous health benefiting volatile essential oils such as *cineol, camphene, borneol, bornyl acetate, α-pinene, etc.*
Chinese teas	Leaves	Improve the immune system, slow down aging and can help prevent cancer. Lower blood, cholesterol and blood pressure and prevent arteriosclerosis.
Ginger	Fresh and dried rhizomes	Use as medicine for vomiting, diarrhea, indigestive heat, accumulation of mucus, coagulation of blood or blood circulation problems(due to drop in blood temperature and thus blood become cold and clumpy.
Cinnamon	Inner bark, Leaves and oil	Anti-Clotting It has an anti-clotting effect on the blood. Also used as arthritis relief and anti-bacterial when added to food, it inhibits bacterial growth and food spoilage, making it a natural food preservative. Also smelling cinnamon boosts cognitive function and memory.
Caraway	Seeds	Caraway is used for digestive problems including heartburn, bloating, gas, loss of appetite, and mild spasms of the stomach and intestines. Caraway oil is also used to help people cough up phlegm..
Cumin	Seeds	Cumin seeds have traditionally been noted to be of benefit to the digestive system, and scientific research is beginning to bear out cumin's age-old reputation.
Anise	Seeds	Anise it is greatly used in the form of lozenges and the seeds have also been used for smoking, to promote expectoration. The volatile oil, mixed with spirits of wine forms the liqueur Anisette.
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Rosemary	Leaves and flowers	The herb parts, especially flower tops contain phenolic anti-oxidant rosmarinic acid as well as numerous health benefiting volatile essential oils such as *cineol, camphene, borneol, bornyl acetate, α-pinene, etc.*
Chinese teas	Leaves	Improve the immune system, slow down aging and can help prevent cancer. Lower blood, cholesterol and blood pressure and prevent arteriosclerosis Prevent tooth decay, freshen the breathe and assist in digestion and also Enhance the eliminating functions of the kidneys.
Ginger	Fresh and dried rhizomes	Use as medicine for vomiting, diarrhea, indigestive heat, accumulation of mucus, coagulation of blood or blood circulation problems.
Cinnamon	Inner bark, Leaves and oil	Anti-Clotting It has an anti-clotting effect on the blood. Also used as arthritis relief and anti-bacterial when added to food, it inhibits bacterial growth and food spoilage.

Table (2). *The mean and SD of pesticides Residues* (mg/kg) *in some medicinal plants collected from local marked in Jeddah, Saudi Arabia.*

Pesticides	Investigated Medicinal plants							
Residues	Caraway	Cumin	Anise	Rosemary	Cinnamon	Tea	Sage	Ginger
Malathion	0.3 ± 0.021		0.4 ± 0.001	<LOQ			<LOQ	
Chlorpyrifos	0.03 ± 0.001	0.01 ± 0.002	0.02 ± 0.002		<LOQ	0.01 ± 0.001	0.01 ± 0.001	
Profenofos	0.16 ± 0.003	3.6 ± 0.001	<LOQ					
Pirimiphos-methyl			0.05 ± 0.003	0.19 ± 0.001			± 0.002	
Chlorenapur			0.01 ± 0.002			0.012 ± 0.002		
Chlrofenapur			0.01± 0.001					
Difenoconazole			<LOQ					
Pyridaben						0.04 ± 0.002		
Lmidacloprid						0.04 ± 0.005		
Buprofezin						0.16 ± 0.002		
Acetamiprid						2.01 ± 0.003		
Flusilazole	0.01 ± 0.003	0.03 ± 0.002						
Carbendazim		0.14 ± 0.003						
Metalaxyl		0.08 ± 0.002						
Thiophanate-methyl	0.21 ± 0.004	0.67 ± 0.005						
Thiamethoxam		0.04 ± 0.004						
Acetamiprid	0.06 ± 0.001	0.26 ± 0.001						
Azoxystrobin		0.07 ± 0.003						
Ethion		0.04 ± 0.004						
Lmidacloprid		0.01 ± 0.005						
Ticyclazole	0.06 ± 0.002	0.22 ± 0.002						
Iprobefos		0.06 ± 0.003						
Flusilazole	0.09 ± 0.003	0.03 ± 0.001						
Cypermethrin		0.65 ± 0.004						
Pendimethalin	0.01 ± 0.003	<LOQ						
Pirimiphos-methyl		± 0.002						
Iprodione		0.05 ± 0.003						
Kresoxim-methyl		0.01± 0.005						
Lambda-Cyhalothrin		0.14 ± 0.002						
Propiconazol	0.01 ± 0.004	0.03 ± 0.001						
Tebuconaole	0.06 ± 0.002	0.12 ± 0.004						

3. Results and Discussion

The results of pesticides residues investigation in eight medicinal plants commonly used by peoples namely; (Rosemary; *Rosmarinus officinals* L. & Sage; *Salvia officinals* L.), family Lamiaceae, (Anise; *Pimpinella anisum* L., Caraway , *Carum carvi* L. & Cumin ; *Cuminum cyminum* L.) family Apiaceae , (Cinnamon; *Cinnamomum verum* L.) family Lauraceae, (Ginger; *Zingiber officinale* Roscoe.) family Zingiberaceae and (Tea ; *Camellia sinensis* L.) family Theaceae. Represented in tables (1&2) showed that all tested medicinal plants except Ginger samples had some detectable pesticides residues (Table 2).

It was found that Ginger samples were free from any pesticides residues ,followed by Cinnamon samples which showed only level of chloropyrifos in level less than the detectable <LOQ while Cumin and Caraway samples showed the highs levels of both pesticides and fungicides as compared to other investigated plant samples. On the other hands Anise contain only difenoconazole which was in level <LOQ The detected fungicides in Cumin and Caraway samples was in form of Azoles compounds (tebuconazole, propiconazole , flucilazole, difenoconazole), carbamate compounds (carbendazim) and other fungicides (iprodione, azoxystrobin, metalaxyl, flusilazole, thiophanate-methyl, ticyclazole, kresoxim-methyl and pendimethalin). Insecticides were mainly predominant in Cumin, Caraway, Anise, Tea, Rosemary, and Sage samples. Insecticides were mainly organophosphates (malathion, chlorpyrifos, profenofos, pirimiphosmethyl, ethion.

The medicinal plants have been looked at in a positive way as they treat many diseases and their active ingredients are active principles of many pharmaceutical compounds that are more safe when compared with the synthetic chemical medication as they might have no or low side effects. From this point of view the consumption of these plants are dramatically increasing specially with the high cost effects of prescription drug in developing countries and according to increasing consumption their economic values and in turn increasing the land implemented by these plants also increased [17].

Pesticides levels in different investigated samples were ranged between non detected and highly presented as in case of Ginger and Cumin samples.

Differences if assumed the same kinds of pesticides were used in both plants might be attributed to part of plant used (rhizomatic roots in Ginger and seeds in Cumin) as well as structure of the used part as in case of Ginger where the moisture content of rhizomes is more that 80% compared to 6% in Cumin seeds as well as high volatile oil contents of Cumin seeds compared to Ginger [15]. On the other hands oils might play a vital role in the solubility of many pesticides in Cumin seeds as compared to high moisture content in Ginger assuming many of pesticides are oil soluble compared to water. Currently, there is considerable variations from country to country in the quality control of levels of pesticides in commodity these variations not only impacts on public health, as contaminants in herbs may represent avoidable risks for patients and consumers, but also has effects on international trade.

Since the 1940s, the amount of synthetic chemical pesticides used annually worldwide has increased, resulting in considerable human health hazards. Malnutrition and infectious diseases often intensify the negative effects of pesticide poisoning. Pesticide health hazards may be in the form of acute effect with symptoms of fatigue, headaches and body aches, skin discomfort, skin rashes, poor concentration, feelings of weakness, circulatory problems, dizziness [10].

Symptoms are often diffuse or do not become apparent for a long time, which then leads to late effects. Many pesticides that are commonly used today are possibly or probably carcinogenic for humans. Other studies have revealed a correlation between pesticide use and sarcomas, multiple myelomas, cancer of the prostate, pancreas, lungs, ovaries, the breasts, testicles, liver, kidneys ,and intestines as well as brain tumors [16]. Furthermore pesticides can damage the human nervous system. Various studies have also shown that the risk of developing Parkinson's disease can increase if there has been contact with specific pesticides.

4. Conclusion

The medicinal plants and natural herbs must be used after the application of safety experiments and safety tests. It is recommended that every country produce medicinal plants and spices (collected from natural habitats or cultivated) should have at least one control laboratory capable of performing the determination of pesticides residues in these plants and give a license for safety to be used in safe ways. Also it is recommended that the medicinal plants must be cultivated without any chemical treatments and stored at good conditions without insecticides treatments.

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