Multi-residue Pesticides in Conventional and Organic Orange

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Authors’ contributions

This work was carried out in collaboration between all authors. Author MHES designed the study, wrote the protocol and supervised the work. Authors MHES, MTS and SNAN carried out all laboratories work and performed the statistical analysis. Authors MTS and MHES managed the analyses of the study. Authors MHES, MTS and SNAN wrote the first draft of the manuscript, managed the literature searches and edited the final manuscript. All authors read and approved the final manuscript.

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ABSTRACT

A monitoring project for pesticide residues in orange samples in Al-Tamer vegetables market, Riyadh was conducted. A total of 144 samples of organic and non-organic orange were collected according to the Codex Alimentarius recommendations. Samples were extracted with acetonitrile and subjected for clean-up using Florisil column. Clean extracts were analyzed using GC-MS against 86 pesticides of concern. Method performance parameters are reported. Organic orange

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samples of both countries contained non-detectable amounts of the tested pesticides. For the non-organic produce samples it contained varying amounts of pesticides depending on the season, country and month. Mostly, insecticides were detected mostly in the samples then fungicides. Violating pesticides were also the anti-insect ones. Methomyl was the violating compound in South African orange and ethion in Egyptian one. Fungicides were below the corresponding MRL. Total amount of residues were the maximum in December 2010 (5.16 mg/kg) followed by November 2010 (4.27 mg/kg) of which ethion was the major constituent, this may be due to insect control practices. The highest level of residues appeared in the November 2011 with 1.68 mg/kg residues of Methomyl followed by august 2011 with 1.3m mg/kg consist of Methomyl and Chlorpyrifos-methyl.

Keywords: GC-MS; pesticide residues monitoring; multi-residues analysis; orange.

1. INTRODUCTION

Increased demands are being placed on farmers to produce food more efficiently and safely. Food safety is a major public concern worldwide. The major pathway of human exposure to pesticides is food ingestion followed by inhalation and dermal contact [1]. Freshly consumed Vegetables and fruits, are of high concern, as they do not receive any cooking treatments, increasing the problem of pesticide residues [2]. A risk assessment is necessary to ascertain the health effects due to intake of pesticide residues in food. Pesticides are used during production and post-harvest treatment of agricultural commodities to guarantee higher productivity of crops and better quality of food [3]. Due to the poor pesticide handling practices and use of more toxic pesticides, inadequate management and regulation of these chemicals in developing countries [4] pesticide contamination levels are higher in the developing countries than the developed ones [5]. Either control programs for pesticide residues in poor countries are limited for lack of resources or proper legislations are not set. Many farmers do not observe the set Pre-Harvest Interval (PHI) because of their high demand for farm produce and low perception of the toxic effects of pesticide residues in food [6]. Increased use of chemical pesticides has resulted in occurrence of residues in food commodities [7] and contamination of the environment [8]. Pesticide residues have been linked with a wide spectrum of human health hazards, ranging from short-term impacts such as headaches and nausea to chronic impacts like cancer, reproductive harm, and endocrine disruption [9].

Pesticide residue monitoring is the only tool to control the quantity of pesticides on food. For the past few decades regulatory authorities in many countries have been setting up monitoring systems for the agricultural products and the environment. The surveillance focuses on the proper use of pesticides in terms of authorization and registration (application rates and pre-harvested intervals), and on compliance with maximum residue limits (MRLs). Pesticide residue monitoring is also recognized as a significant aspect of initiatives to reduce potential hazards to human health [10-13].

Fruits and vegetables have been given a lot of attention in monitoring programs since most of them are eaten raw, it is expected that they contain higher pesticide residue levels compared to other food groups of plant origin. According to the Pesticide Residues Committee in the UK, consumers are encouraged to eat at least five portions of fruits and vegetables daily. Therefore, assessing the risk of pesticide residues in these commodities intended for human consumption is necessary [14-17].

This study aimed to monitoring the levels of pesticide residues in conventional and organic oranges imported from Egypt and South Africa. Pesticide residues in terms of type and quantity in orange fruits imported from Egypt and South Africa as two major producing and supplying countries of Saudi Market. The study targeted Navel orange produced organically or by conventional farming.

2. MATERIALS AND METHODS

2.1 Sampling

Samples were collected from Al-Tamer Market, Riyadh, Saudi Arabia. Egyptian Orange (Navel) samples were collected during the active production season months from November 2009 to February 2010 and from November 2010 to February 2011. In case of South African orange (Navel), samples were collected during the active
production season from July to October 2010 and from July to October 2011. Samples were collected from non-organic orange (conventional agriculture product) and organically produced orange. Codex Alimentarius procedures were followed in sampling and transporting. Samples were collected monthly and were kept in polyethylene bags in fridge at 4°C until analyzed.

2.2 Sample Preparation

The chopped orange subsamples (100 g) were placed in a stainless steel jar 1 L and extracted with 200 ml of acetonitrile and 10 g celite, the mixture was vigorously homogenized at high speed for 2 min. and the mixture was filtrated using Buchner funnel fitted with shark-skin filter paper into 50 ml suction flask. Organic extract was transferred to 1L separator funnel and 100 ml of (PE) was added, the mixture was vigorously shaken for 2 min and then 100 ml saturated solution of NaCl and 600 ml water were added. The mixture was vigorously mixed and the separator funnel was allowed to be held at horizontal position for few minutes. The aqueous layer was discarded and the solvent layer was washed twice with 100 ml portions of distilled water and the washed layer was transferred into 100 ml beaker and 15 g of anhydrous sodium sulphate was added and filtered through Whatman filter paper. Finally the extract was concentrated to 5ml in rotary evaporator and transferred directly to florisil column for clean-up. Column was prepared according to the AOAC method (1995, chap. 10). The column was contained about 12 cm activated florisil topped with 1 cm anhydrous sodium sulphate, column was washed by 40 ml (PE) and then sample was added and allowed to pass through the column. The walls of the tube were rinsed additional small portions of petroleum ether and elute at 5 ml/min with 200 ml 6% eluting solvent (Diethyl ether in (PE)), and then 200 ml 15% and finally 200 ml 50% eluting solvent (Diethyl ether in (PE)) at 5 ml/min. column effluent was collected, solvent was evaporated and residue was transferred to injection vials then subjected to determination step.

2.3 Reagents and Equipment

All pesticides standard were obtained from (Riedel de Haen and Supelco). 1 mg/ml stock solution of each was prepared by dissolving 20 mg of the pure analytical standard in acetone. A single composite standard solution was prepared by diluting with acetone according to limit of detection (LOD). All standard solutions were stored in glass-Stoppard flasks at 4°C. Mixed compound calibration solutions were prepared in acetone and they were used as spiking solution. Solvents (residue analysis grade) used were acetone, acetonitrile, petroleum ether and other reagents such as sodium chloride and anhydrous sodium sulphate, florisil 60-100 mesh for residue analysis were also from purchased from (Fluka). The florisil and anhydrous sodium sulphate was activated at 100°C overnight and stored in 500 ml glass flask with glass stoppers and stored in oven at 100°C. The equipment used were a high-speed blender with a stainless steel jar (Waring, USA), a separation funnel shaker (GFL, Germany), a rotary evaporator, R 215 with cooler circulator chiler B-740 (Buchii, Switzerland), Buchner funnel and chromatographic tubes with Teflon stopcocks and course fritted glass (Agilent, USA) and syringes (Hamilton Bonadus AG, Switzerland). All glassware were rinsed thoroughly using soap and deionization water, then washed with acetone and dried in oven (100-130°C) over night.

2.4 Instrumentation

Gas chromatograph-mass spectrometer (Agilent model 6890N) coupled with (model 5975B) quadruple mass spectrometer with a GC column HP-5MS 5% phenyl - 95% methyl siloxane, 30m x 0.25 mm id x 0.25 µm film thickness was used (plate 3). GC operating conditions: splitless injection, injector temperature 250°C, helium carrier gas (99.9999 purity) at flow rate 0.9 ml/min with column head pressure 7.4 psi, oven temperature from 70°C (2 min hold), than raised to 130°C at the rate (25°C/min) afterwards raised to 220°C at (2°C/min) and then raised to 280°C at (10°C/min) and eventually (4.6 min hold). The sample (1 µL) was injected in splitless modes. The MS system was routinely set in selective ion monitoring (SIM) mode and each compound was quantified based on peak area using one target ion (quantifier) and one or two qualifying ion. Mass spectrometer parameter was set as follows: Electron impact ionization mode with 70 eV electron energy, scan mass range 100-400 at 0.62 sec/cycle. Ion source temperature 230°C, MS quad temperature 150°C, EM voltage 1450 and solvent delay time is 4 min.

2.5 Determination of Pesticide Residues

An appropriate aliquot (2 µl) of samples was injected in Gas Chromatography Mass spectrometry under the mentioned conditions and then pesticides residues had been identified by comparison of retention time values with
reference standard. Confirmation of results was performed using selected ion monitoring (SIM) mode, one is the quantifier and the qualifying ion. Table 3 shows the retention time and characteristic masses (m/z) for each pesticide.

### 2.6 Recovery

Orange samples, free of tested pesticides, were used for the fortification experiments. 100 g homogenized sample was spiked prior to determination procedure by addition of a mixture of standard pesticides solution 0.1 µg/ml of each compound. Spiked samples were left to stand for 30 min. to allow pesticide to absorb into the sample. Samples were extracted according to the method described above. The limit of detection (LOD) and recoveries data for tested pesticides are tabulated in Table 1.

| Compounds         | Retention time (min) | LOQ | Recovery % | Target ion m/z | Qualifier ions m/z |
|-------------------|----------------------|-----|------------|----------------|-------------------|
|                   |                      |     |            |                |                   |
| Dichlorvos        | 7.211                | 0.02| 59         | 64             | 109               |
| Propamocarb       | 9.849                | 0.04| 78         | 96             | 58                |
| Mevinphos         | 10.828               | 0.09| 63         | 49             | 127               |
| Chloroneb         | 13.015               | 0.01| 79         | 77             | 191               |
| Methomyl          | 14.837               | 0.01| 44         | 53             | 105               |
| Propachlor        | 16.350               | 0.02| 65         | 96             | 120               |
| Propoxur          | 16.440               | 0.03| 77         | 91             | 110               |
| Ethoprophos       | 17.183               | 0.05| 72         | 64             | 157               |
| Bendiocarb        | 18.808               | 0.01| 63         | 100            | 151               |
| Sulfotep          | 19.350               | 0.01| 50         | 100            | 322               |
| Alfa-BHC          | 19.449               | 0.02| 78         | 95             | 183               |
| hexachlorobenzen  | 19.949               | 0.05| 66         | 75             | 284               |
| Dichloran         | 20.412               | 0.03| 32         | 65             | 176               |
| Dimethoate        | 20.694               | 0.02| 46         | 43             | 87                |
| Simazine          | 21.240               | 0.01| 63         | 98             | 201               |
| Carbofuran        | 21.517               | 0.01| 50         | 72             | 164               |
| Lindan            | 21.953               | 0.01| 58         | 100            | 219               |
| Fonofos           | 22.837               | 0.06| 56         | 79             | 109               |
| Delta-BHC         | 23.934               | 0.05| 91         | 47             | 181               |
| Diazinon          | 24.181               | 0.03| 66         | 36             | 179               |
| Ipobenfos         | 25.472               | 0.04| 47         | 93             | 91                |
| Pirimicarb        | 26.194               | 0.05| 71         | 57             | 166               |
| Dichlorfenthion   | 26.869               | 0.01| 39         | 63             | 279               |
| Phosphamidon I    | 27.074               | 0.03| 80         | 100            | 127               |
| Phosphamidon II   | 27.105               | 0.05| 59         | 66             | 127               |
| Chlorpyrifos-Me   | 27.530               | 0.03| 86         | 71             | 286               |
| Vinclozolin       | 27.643               | 0.02| 36         | 88             | 212               |
| Carbaryl          | 27.846               | 0.03| 59         | 100            | 144               |
| Alachlor          | 28.292               | 0.04| 71         | 63             | 160               |
| Ronnal            | 28.738               | 0.02| 60         | 97             | 285               |
| Metalaxyl         | 28.894               | 0.05| 63         | 102            | 206               |
| Fenitrothion      | 30.004               | 0.07| 45         | 58             | 277               |
| Linuron           | 30.118               | 0.04| 41         | 72             | 61                |
| Aldrin            | 30.417               | 0.02| 57         | 103            | 66                |
| Thiothionic acid  | 30.794               | 0.08| 76         | 49             | 100               |
| Malathion         | 31.310               | 0.03| 81         | 85             | 127               |
| Fenthion          | 31.679               | 0.03| 58         | 100            | 278               |
| Pirimiphos-ethyl  | 34.279               | 0.05| 49         | 93             | 318               |
| Compounds               | Retention time (min) | LOQ     | Recovery % | Target ion m/z | Qualifier ions m/z |
|------------------------|----------------------|---------|------------|----------------|-------------------|
|                        |                      |         |            | Spiked 1       | Spiked 2          | Q₁  | Q₂  |
| Capten                 | 34.791               | 0.06    | 76         | 98             | 79                | 151 | 114 |
| Chlorofenvfenphos      | 35.549               | 0.05    | 78         | 95             | 267               | 323 | 269 |
| Chlorodan-trans        | 35.990               | 0.05    | 64         | 100            | 373               | 375 | 237 |
| Alfa-endosulfan        | 36.919               | 0.03    | 98         | 107            | 239               | 237 | 195 |
| Nanchlor-trans         | 37.168               | 0.02    | 49         | 100            | 409               | 100 | 237 |
| Chlordane-cis          | 37.311               | 0.01    | 77         | 98             | 375               | 373 | 377 |
| Disulfoton sulfur      | 37.670               | 0.01    | 53         | 97             | 213               | 153 | 97  |
| Dieldrin               | 39.172               | 0.02    | 96         | 100            | 79                | 265 | 81  |
| P,P-DDE                | 39.688               | 0.03    | 91         | 90             | 246               | 318 | 248 |
| O,P-DDD                | 40.321               | 0.04    | 64         | 67             | 235               | 237 | 165 |
| Endrin                 | 40.920               | 0.01    | 44         | 95             | 263               | 265 | 281 |
| Beta-endosulfan        | 41.820               | 0.03    | 97         | 63             | 207               | 239 | 195 |
| Chlorobenzilate        | 42.683               | 0.01    | 49         | 84             | 251               | 139 | 253 |
| P,P-DDD                | 43.234               | 0.04    | 91         | 49             | 235               | 237 | 165 |
| Benodanil              | 43.773               | 0.03    | 94         | 73             | 231               | 323 | 203 |
| Ethion                 | 44.131               | 0.03    | 47         | 89             | 231               | 97  | 153 |
| Carbophenothon         | 45.558               | 0.04    | 71         | 37             | 157               | 121 | 125 |
| Resmethrin I           | 48.965               | 0.03    | 65         | 71             | 123               | 171 | 143 |
| Resmethrin II          | 49.532               | 0.04    | 49         | 43             | 123               | 171 | 143 |
| Hexabromobenzen        | 49.772               | 0.02    | 52         | 59             | 551.6             | 554 | 549.6|
| Phosmet                | 50.300               | 0.07    | 118        | 71             | 160               | 161 | 77  |
| EPN                    | 50.726               | 0.02    | 49         | 100            | 157               | 169 | 185 |
| Dicofol                | 50.955               | 0.05    | 97         | 96             | 139               | 111 | 251 |
| Fenoxycarb             | 51.040               | 0.01    | 88         | 93             | 255               | 186 | 116 |
| Tetramethrin II        | 51.322               | 0.04    | 73         | 68             | 164               | 123 | 81  |
| Tetradelton            | 52.029               | 0.03    | 77         | 29             | 159               | 111 | 229 |
| Mirex                  | 52.460               | 0.05    | 50         | 89             | 272               | 274 | 270 |
| Furathiocarb           | 52.620               | 0.03    | 46         | 55             | 163               | 57  | 164 |
| Amitraz                | 53.373               | 0.04    | 62         | 69             | 132               | 121 | 147 |
| Lamma-cyhalothrin      | 53.770               | 0.04    | 73         | 83             | 181               | 197 | 208 |
| Azenophos-ethyl        | 53.882               | 0.04    | 21         | 77             | 132               | 160 | 77 |
| allethrin I            | 54.515               | 0.02    | 60         | 67             | 123               | 181 | 81  |
| allethrin II           | 54.523               | 0.02    | 49         | 102            | 123               | 181 | 81  |
| allethrin III          | 54.590               | 0.02    | 65         | 91             | 123               | 181 | 81  |
| Permethrin I           | 54.891               | 0.05    | 89         | 100            | 183               | 163 | 165 |
| Permethrin II          | 55.111               | 0.04    | 99         | 93             | 183               | 163 | 165 |
| Comaphos               | 55.165               | 0.04    | 101        | 99             | 263               | 226 | 109 |
| Cyfluthrin III         | 55.855               | 0.04    | 77         | 89             | 163               | 165 | 226 |
| Cyfluthrin I           | 55.997               | 0.03    | 94         | 100            | 163               | 165 | 226 |
| Cyfluthrin IV          | 56.103               | 0.03    | 100        | 102            | 163               | 165 | 226 |
| Cyfluthrin II          | 56.162               | 0.03    | 97         | 93             | 163               | 165 | 226 |
| Cypermethrin II        | 56.284               | 0.03    | 87         | 100            | 163               | 165 | 181 |
| Cypermethrin IV        | 56.424               | 0.02    | 84         | 100            | 163               | 165 | 181 |
| Cypermethrin I         | 56.522               | 0.02    | 105        | 97             | 163               | 165 | 181 |
| Cypermethrin III       | 56.575               | 0.05    | 59         | 99             | 163               | 165 | 181 |
| Fenvalerate I          | 57.454               | 0.03    | 66         | 36             | 125               | 167 | 281 |
| Fenvalerate II         | 57.716               | 0.04    | 47         | 93             | 125               | 167 | 281 |
| Deltamethrin           | 58.440               | 0.03    | 87         | 100            | 253               | 181 | 181 |
3. RESULTS AND DISCUSSION

The method used for analysis has been published before [18] and it was checked for performance appropriateness before analysis. Linearity, recovery LOQ and reproducibility parameters were tested.

3.1 Pesticide Residues in Conventional (Non-organic) Orange Samples

3.1.1 Egyptian product

Data in Table 2 summarize average levels of pesticide residues in the collected representative samples of orange imported from Egypt during the active production season from November 2009 to February 2011 for 2 successive seasons. Out of 86 tested pesticides only nine compounds were found i.e. Chlorpyrifos-methyl, Pirimiphos-methyl, Dimethoate, Ethion, Propamocarb, Permethrin, Fenvalerate, Dicofol, Vinclozolin. Detected compounds belong to different pesticide chemical groups, organophosphorus, pyrethroids, organochlorine and dicarboximides. Mostly they possess insecticidal/.acaricidal activity only Vinclozolin in a fungicide, this may be attributed to the fact that insecticides are used much more than fungicides in open field, similar results are obtained by [19]. Fig. 1 is showing the number of violating samples exceeding MRL, [20] that was 5 times for Ethion and 4 times for Dicofol. Chlorpyrifos-methyl, Propamocarb and Vinclozolin were lower than MRL in all samples during the two tested seasons. Samples of February 2009 scored the highest record of detection times of pesticides as 7 pesticides were detected three of them were higher than the MRLs. Total amount of residues were the maximum in December 2010 (5.16 mg/kg) followed by November 2010 (4.27 mg/kg) of which ethion was the major constituent, this may be due to insect control practices as shown in Fig. 2.

3.1.2 South African product

Pesticide residues detected in South African orange are tabulated in Table 3. Samples were collected during the active production season from July 2010 to November 2011. The pesticides detected are mostly insecticides as in the Egyptian Product data above. The detected pesticides were the insecticides; Chlorpyrifos-methyl, primiphos-methyl, dimethoate, methomyl and the fungicides; propamocarb and benodanil. The most violating pesticide was methomyl with 5 records of exceeding limits followed by dimethoate (3 times) and chlorpyrifos-methyl (3 times). Both fungicides were below the limits during both tested seasons as shown in Fig. 3. The highest level of residues appeared in the

| Detected pesticides | November (2009) | December (2010) | January (2010) | February (2011) | MRL | V |
|---------------------|-----------------|-----------------|----------------|-----------------|-----|--|
| Chlorpyrifos-methyl | nd               | nd              | 0.667          | 0.017           | 0.302 | 0.036 | 0.38 | 0.002 | 0.50 | 0 |
| Pirimiphos-methyl   | nd               | 0.644           | 0.179          | nd              | 1.146* | nd | 0.185 | 0.012 | 1.00 | 1 |
| Dimethoate          | 1.06*            | nd              | 0.173*         | nd              | nd | 0.076* | nd | 0.035* | 0.02 | 3 |
| Ethion              | nd               | 3.581*          | 0.542*         | 3.666*          | nd | 0.176* | nd | 0.035* | 0.01 | 5 |
| Propamocarb         | nd               | nd              | nd             | 0.004           | nd | 0.056 | nd | 0.10 | nd | 0 |
| Permethrin          | Nd               | 0.045           | 0.014          | nd              | nd | 0.742* | 0.742* | 0.05 | 2 |
| Fenvalerate         | 0.0586*          | nd              | 0.0086         | 0.065*          | 0.176* | nd | nd | 0.02 | 3 |
| Dicofol             | 0.173*           | nd              | 1.306*         | nd              | 2.146* | 1.435* | 0.02 | 4 |
| Vinclozolin         | Nd               | 0.0033          | 0.0233         | nd              | nd | 0.0133 | nd | 0.05 | 0 |
| No. of detections   | 3                | 3               | 5              | 5               | 7 | 5 |
| Total residues     | 1.29             | 4.27            | 1.40           | 5.16            | 1.53 | 0.388 | 3.59 | 2.226 | |

Each figure is an average of 3 replicates
nd: not detected
*violating sample
V: number of violating samples
November 2011 with 1.68 mg/kg residues of methomyl followed by August 2011 with 1.3 mg/kg consist of methomyl and chlorpyrifos-methyl (Fig. 4). July 2011 and October 2011 samples have 3 detection records. All samples contained pesticides residues at least once.

Fig. 1. Number of violating Egyptian orange samples of each detected pesticides in the whole study samples

Fig. 2. Comparison of total pesticide residues found in each month in both seasons of the study in Egyptian orange

Fig. 3. Number of violating South Africa orange samples of each detected pesticides in the whole study samples
Table 3. Pesticides residues detected (mg/kg) in non-organic orange imported from South Africa (2010-2011)

| Detected pesticides          | July (2010) | August (2010) | September (2010) | October (2010) | November (2010) | MRL V | July (2011) | August (2011) | September (2011) | October (2011) | November (2011) | MRL V |
|------------------------------|-------------|---------------|------------------|----------------|----------------|-------|-------------|---------------|------------------|----------------|----------------|-------|
| Chlorpyrifos-methyl          | 0.026       | 0.185*        | nd               | 0.666*         | 0.055*         | nd    | 0.055*      | 3             |                  |                |                | 0.50  |
| Pirimiphos- methyl           | 0.0123      | nd            | nd               | 0.066          | nd             | 0.379 | nd          | nd            |                  |                |                | 1.00  |
| Dimethoate                   | nd          | 0.38*         | nd               | 0.076*         | nd             | 0.326*| nd          | 3             |                  |                |                | 0.379 |
| methomyl                     | nd          | 1.180*        | 0.665*           | 0.433*         | 0.073*         | 1.681*| 0.073*      | 0.02          | 1.681*           | 0.02          | 5               |
| Propamocarb                  | nd          | nd            | nd               | 0.007          | nd             | 0.073*| nd          | 0.02          | 0.073*           | 0.02          | 1.681*          |
| benodanil                    | nd          | nd            | nd               | nd             | 0.007          | nd    | 0.007       | 1             | 0.007            | 0.01          | 0.50  |
| No. of detections            | 2           | 3             | 2                | 2              | 1              | 2     | 2           | 3             | 3                | 1            | 1               |
| Total residues detected (ppm)| 0.037       | 0.418         | 1.256            | 1.332          | 0.066          | 0.440 | 0.062       | 0.649         | 0.379            | 1.681         |

Each figure is an average of 3 replicates
nd: not detected
*violating sample
V: number of violating samples

Fig. 4. Comparison of total pesticide residues found in each month in both seasons of the study in South Africa orange

3.2 Pesticide Residues in Organically Produced Orange

Fortunately, all organic samples collected from the Egyptian or the South African products were free of tested pesticides to the limit of quantification of the method reported above (Table 1). These results are very much encouraging for consuming organic orange from both countries, with taking in consideration the price issue.

4. CONCLUSIONS

A total of 144 samples of organic and non-organic orange samples collected for 2 seasons from Saudi market imported from Egypt and South Africa. Organic orange samples of both countries contained non-detectable amounts of the analyzed pesticide residues. For the non-organic produce samples it contained varying amounts of pesticides depending on the season, country and month. Mostly, insecticides were found mostly in the samples then fungicides. Violating pesticides were also the anti-insect ones. Methomyl was the violating compound in South African orange and Ethion in Egyptian one. Fungicides were below the corresponding MRL. The number of detected pesticides in Egyptian orange (9) was higher than South African product (6). Total residues in Egyptian orange were higher than the product of South Africa.
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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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