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Insecticidal properties of plant extracts and monoterpenes towards the fourth instars of *Spodoptera littoralis* Boisd (Lepidoptera: Noctuidae) and adults of *Aphis fabae* L. (Hemiptera: Aphididae)

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ABSTRACT

The extracts of three plants, namely *Zygophyllum coccineum* L., *Majorana hortensis* (Moench) and *Mentha microphylla* L. were tested for their insecticidal activity against the fourth instar larvae of the Egyptian cotton leafworm, *Spodoptera littoralis* and the adults of the bean aphid, *Aphis fabae*. All of the tested extracts were found to be toxic against both insects. However, the degree of toxicity was varied based on plant extract, plant species and insect species. Among the tested extracts, petroleum ether extract of *M. hortensis* (LD_{50} = 2.76 µg/larva) was the most potent against *S. littoralis* followed by chloroform extract of the same plant (LD_{50} = 3.36 µg/larva). The petroleum ether and ethanol extracts of *Z. coccineum* showed the less activity against *S. littoralis*. In the case of *A. fabae*, the petroleum ether and chloroform extracts of *M. hortensis*, the petroleum ether and ethanol extracts of *M. microphylla* showed pronounced toxic effect with LC_{50} values of 2.21, 2.44, 2.62 and 2.25 g/l, respectively. The essential oil of *M. hortensis* was isolated by hydrodistillation and analyzed by gas chromatography-mass spectrometry (GC-MS). Terpinen-4-ol (30.0%), γ-terpinene (11.3%), and *trans*-sabinene hydrate (10.8%) were the major components of the oil. This oil revealed remarkable insecticidal activity against *S. littoralis* and *A. fabae*. Repetitive column chromatography of the essential oil of *M. hortensis* led to isolate the two major constituents (terpinen-4-ol and γ-terpinene). The structure of these compounds were determined based on their specral data of ^{1}H NMR, ^{13}C NMR and MS. γ-Terpinene was more toxic than terpinen-4-ol towards *S. littoralis* and *A. fabae*. The isolated compounds were less toxic than the essential oil against both insects.

Key words: Plant extracts, *Majorana hortensis*, essential oil, monoterpenes, insecticidal activity, *Spodoptera littoralis*, *Aphis fabae*.

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INTRODUCTION

The control of pests has become increasingly difficult because of reduced effectiveness of pesticides caused by emergence of pesticidal resistance in arthropod pests. Therefore, an effort is warranted to find alternatives or formulations for improving currently used pesticides. The search for new natural products with promising activity continues in academia and in industry, to initiate and to foster novel, ecologically and economically sound crop protection solutions. The majority of botanicals that were developed as commercial pesticides originated from tropical and subtropical sources. Because of the intensity of plant-pest interactions in tropical and subtropical regions, the plants in these regions have well-developed defense mechanisms against pests and are excellent sources of new pesticidal substances (Prakash and Rao, 2000).

Zygophyllum coccineum L. (Zygophyllaceae) grows wild in Egypt and in the neighbouring region of Sudan. Leaves, stems and fruits of this plant are used in folk medicine as the drug against rheumatism, gout, asthma and hypertension and are also used as a diuretic, anthelminthic and antidiabetic agents. Some chemical constituents, such as quinovic acid, saponins, triterpenoid saponins and tannins have been found in the leaves, stems and roots of *Z. coccineum* (Elgamal *et al.*, 1995). *Majorana hortensis* Moench (Lamiaceae), is an herbaceous, perennial plant, native to the Mediterranean basin. It is cultivated not only in Mediterranean countries, but also in Central and Eastern Europe (Novak *et al.*, 2000). Marjoram is mainly used as a spice in the food industry (Burdock, 1995). The plant is also medicinally valuable because of its stimulant and antispasmodic properties, and it is a good general tonic, treating various disorders of the digestive and respiratory systems (Chevallier, 1996). *Mentha microphylla* L. (Lamiaceae) is a perennial herb commonly growing on the sides of irrigation canals and bounds in most of cultivated areas in Egypt. The plant is commonly used in folk medicine as carminative, antiseptic and stimulant (Batanouny, 1999).

The cotton leafworm, *Spodoptera littoralis* Boisduval (Lepidoptera: Noctuidae), and the bean aphid, *Aphis fabae* L. (Hemiptera: Aphididae), are considered two of the most destructive insect pests of a variety of crops in many areas of the world (Abdelgaleil *et al.*, 2002; Ahmed *et al.*, 2007). In this work, the potential insecticidal activity of three Egyptian plant extracts and *M. hortensis* essential oil on the fourth instar larvae of *S. littoralis* and the adults of *A. fabae* were presented. In addition, the chemical composition

of *M. hortensis* essential oil was determined by using GC/MS. The oil major constituents were isolated and identified, and their insecticidal properties of the isolated compounds were also evaluated.

MATERIALS AND METHODS

General analytical and experimental procedures: Solvents were obtained from Algoumhoria Chemical Company (Alexandria, Egypt). The solvents were the highest purity available and were distilled before using.Nuclear magnetic resonance (NMR) spectra were recorded in $CDCl_3$ on a JEOL JNM ECD 500 Spectrometer operating at 500 MHz for ^1H and 125 MHz for ^{13}C. Infrared (IR) spectra (KBr) were measured on a Perkin-Elmer 1430 spectrophotometer. Ultraviolet (UV) spectra were recorded in MeOH on a HEλIOSα spectrophotometer. Mass spectra were measured on JEOL JMS-AX500 mass spectrometer. Silica gel 70-230 mesh (Merck) was used on open column chromatography. The GC/MS analysis of the essential oil was conducted on a gas chromatograph (TRACE GC 2000: THERMO, Cairo, Egypt)/mass spectrometer (SSQ 7000: FINNIGAN, Cairo, Egypt).

Insects: A laboratory strain of *S. littoralis* was obtained from the Bioassay Laboratory, Faculty of Agriculture, Alexandria University. The colony was reared under laboratory conditions on caster bean leaves, *Ricinus communis* L. (Euphorbiaceae), at 26 ±2 °C and 70 ± 5% r.h. (Eldefrawi *et al.*, 1964). A field strain *A. fabae* was collected from a farm in Elbehera Governorate, north Egypt, and reared under laboratory conditions on faba bean plants at 22-25°C and 65 ± 5% r.h. (Norman and Sutton, 1967).

Plant materials: The leaves of *Zygophyllum coccineum* L. and *Majorana hortensis* (Moench) were collected from Sinai Peninsula desert, Egypt and *Mentha microphylla* L. leaves were collected from a farm close to Damanhour City, Egypt in September, 2007. The plant samples were identified by Prof. Dr. Fath Allah Zitoon of Alexandria University. Voucher specimens have been deposited in the herbarium of the Faculty of Agriculture, Alexandria University.

Preparation of plant extracts: Leaves of the three tested plants were washed and air-dried at room temperature (26 ± 2 °C) for two weeks. The plant samples (250 g) were ground and extracted with petroleum ether (60-80 °C), chloroform and ethanol (one liter of each), successively, in a shoxhlet apparatus for four hours. The solvents were evaporated under

vacuum and the resulting extracts were evaluated for their insecticidal activities.

Isolation and analysis of *Majorana hortensis* essential oil: Essential oil of air-dried leaves of *M. hortensis* was extracted by hydrodistillation in a Clevenger-type apparatus for 2 h (Guenther, 1952). The oil was dried over anhydrous sodium sulfate. The essential oil was diluted in diethyl ether and 1 µl was injected into a gas chromatograph/mass spectrometer. The GC column was a DB-5 (5%-Phenyl, methylpolysiloxane) capillary column (60 m × 0.25 mm i.d.). The GC conditions were as follows: injector temperature, 220°C; column temperature, isothermal at 40°C for 2 min, then programmed to 250°C at 5°C/2 min and held isothermal for 2 min; ion source temperature, 200°C. Helium was used as a carrier gas at a rate of 1 ml/min. The effluent of the GC column was introduced directly into the ion source of the MS. Spectra were obtained in the EI mode with 70 eV ionization energy. The sector mass analyzer was set to scan from 40 to 400 amu for 5 s.

Isolation and identification of major constituents of *Majorana hortensis* oil: The essential oil of *M. hortensis* (10 g) was chromatographed on silica gel (250 g) using a hexane/acetone solvent system, starting with fractions of 100 ml of hexane (10×), 2% acetone/hexane (20×), 5% acetone/hexane (10×), 10% acetone/hexane (5×), and finally with acetone (5×) (Abbassy *et al.*, 2009). The resulting fractions were pooled to two main fractions based on their TLC profiles. The first main fraction (fractions 20-26, 4.1 g) was further purified on a silica gel column eluted with 2% acetone/hexane to give 2.3 g of γ-terpinene. Similarly, the second main fraction (fractions 38-40, 8.1 g) was subjected to a silica gel column eluted with 10% acetone/hexane to offer 4.3 g of terpinen-4-ol. The structures of γ-terpinene and terpinen-4-ol (Figure 1) were determined by spectral data of ^{1}H-NMR, ^{13}C-NMR and MS.

Bioassay against *S. littoralis*: Topical application technique was used to assess the larvicidal activity of the plant extracts of *Z. coccineum*, *M. hortensis* and *M. microphylla*, and the essential oil *M. hortensis*, and the isolated compounds γ-terpinene and terpinen-4-ol on the fourth instars of *S. littoralis*. Serial concentrations of the plant extracts, the oil, the isolated compounds and a reference insecticide, profenofos, were prepared in acetone. One microliter of test solution was applied on the dorsum of larvae by a microapplicator. The plant extracts and the oil were tested at doses of 1.0, 2.0, 4.0, 6.0, 8.0 and 10.0 µg/larva. γ-Terpinene and terpinen-4-ol were

tested at doses of 5, 10, 20, 30, 40 and 50 µg/larva. Profenofos was tested at concentrations of 0.1, 0.2, 0.3, 0.5, 0.7 and 1.0 µg/larva. Three replicates of 10 larvae were used for each dose and control treatments. The treated larvae were then transferred to glass cups and supplied with fresh castor bean leaves. Mortality (%) was recorded at 24 h post treatment. The lethal doses causing 50% mortality (LD_{50}) expressed as µg/larva were calculated from log-dose mortality regression lines (Finney, 1971).

Bioassay against the adults of *A. fabae* : The insecticidal activity of plant extracts, *M. hortensis* oil, γ-terpinene and terpinen-4-ol was evaluated on the adults of *A. fabae* by using rapid dipping assay. The tested materials were prepared first in acetone. Serial dilutions of these solutions were prepared with distilled water containing 0.05% of a wetting agent (Triton-X 100) to give the required concentrations. The plant extracts were tested at concentrations of 1, 2, 3, 4 and 5 g/l, while the *M. hortensis* oil and the isolated compounds γ-terpinene and terpinen-4-ol were tested at concentrations of 0.5, 1.5, 2.0, 3.0 and 4.0 g/l. Ten aphids were placed in a short glass cylinder with the bottom covered with gauze and internally coated with fluon. The aphids were immersed by placing the cylinder in a shallow Petri dish containing 2 ml of test compound solution for 10 s. Three replicates of each concentration and control were used. Methomyl was used as a reference insecticide. Mortality (%) was recorded after 24 h and LC_{50} was calculated from log-concentration mortality regression lines.

Statistical analysis: Mortality of each dose and/or concentration was calculated after 24 h of treatment as mean of the three replicates with 10 insects each. The mortality data were subjected to Probit analysis (Finney, 1971) to obtain the LD_{50} and LC_{50} values, using the SPSS 12.0. The values of LD_{50} and LC_{50} were considered significantly different, if the 95% confidence limits did not overlap.

RESULTS AND DISCUSSION

Insecticidal activity of plant extracts against *S. littoralis*: The insecticidal activity of the extracts of *Z. coccineum*, *M. hortensis* and *M. microphylla* was evaluated against the fourth instar of *S. littoralis* by using topical application assay. The LD_{50} values, 95% confidence limits and other data generated from regression lines are shown in (Table 1). All of the tested extracts revealed pronounced toxic effect. However, the degree of toxicity was differed based on the solvent and plant. The extracts of petroleum ether

and chloroform of *M. hortensis* and the extracts of petroleum ether and ethanol of *M. microphylla* exhibited the highest toxicity, as their LD_{50} values were 2.76, 3.36, 4.40 and 4.61 µg/larva, respectively. The extracts of petroleum ether and ethanol of *Z. coccineum* showed the less toxicity among the tested extracts.

Table 1: Insecticidal activity of plant extracts against *Spodoptera littoralis* treated by topical application.

Plant	Extract	LD_{50} (µg/larva)	95% confidence limits	Slope
Zygophyllum coccineum	Petroleum ether	8.98	7.23-17.97	4.45±0.54
	Chloroform	5.58	3.45-9.25	2.79±0.27
	Ethanol	6.83	3.04-19.64	4.23±0.47
Majorana hortensis	Petroleum ether	2.76	1.24-3.85	2.89±0.27
	Chloroform	3.36	1.60-4.72	2.86±0.26
	Ethanol	5.02	3.25-7.25	2.71±0.26
Mentha microphylla	Petroleum ether	4.40	2.32-6.53	3.24±0.27
	Chloroform	5.19	3.16-8.11	2.62±0.26
	Ethanol	4.61	2.01-7.61	3.24±0.27

The insecticidal activity of some plant extracts against the larvae of *S. littoralis* has been previously described (Abdelgaleil, 1995; Sadek, 2003; Pavela, 2004; Pavela and Chermenskaya, 2004; Abdel-Rahman and Al-Mozini, 2007). It is difficult to compare the toxicity of these extracts with the tested extracts of the present study due to the difference in bioassay and larval stage. However, the tested extracts were more effective against the larvae of *S. littoralis* than the extracts of *Piper nigrum*, *Datura stromonium* and *Simmondsia chinensis* (El-Gayar *et al.*, 1979; El-Doksh *et al.*, 1984; Abdel-Rasoul, 2006). On contrary, these extracts were less toxic than the acetone and ethanol extracts of *Pancratium maritimum* (Abbassy *et al.*, 1998).

Insecticidal activity of plant extracts against *A. fabae*: The insecticidal activity of the nine tested extracts against the adults of *A. fabae* was summarized in (Table 2). In general, the extracts of *M. hortensis* were more effective than the extracts of *M. microphylla* and *Z. coccineum*. Petroleum ether extract of *M. hortensis* (LC_{50} = 2.21 g/l) and ethanol extract of *M. microphylla* (LC_{50} = 2.25 g/l) showed the highest toxic effect without significant differences. Petroleum ether and ethanol extracts of *Z. coccineum* were the less effective ones among the tested extracts with LC_{50} values of

4.50 and 3.95 g/l, respectively. The toxicity of the tested extracts against the adults of *A. fabae* was higher than that of the extracts of *Lupinus termis*, *L. luteus* and *P. maritimum* (Abbassy *et al.*, 1998; Abbassy *et al.*, 2002).

Table 2: Insecticidal activity of plant extracts against *Aphis fabae* treated by rapid dipping.

Plant	Extract	LC_{50} (g/l)	95% confidence limits	Slope
Zygophyllum coccineum	Petroleum ether	4.50	3.24-13.46	2.93 ± 0.32
	Chloroform	3.18	2.22-5.25	2.93 ± 0.28
	Ethanol	3.95	3.55-4.50	2.79 ± 0.31
Majorana hortensis	Petroleum ether	2.21	0.62-2.86	5.17 ± 0.56
	Chloroform	2.44	1.38-3.05	4.71 ± 0.51
	Ethanol	2.67	2.03-3.45	3.18 ± 0.28
Mentha microphylla	Petroleum ether	2.62	1.48-3.32	5.05 ± 0.51
	Chloroform	3.33	2.65-4.44	3.20 ± 0.31
	Ethanol	2.25	0.44-4.49	2.68 ± 0.28

Extraction and chemical composition of *M. hortensis* essential oil: The essential oil from leaves of *M. hortensis* was isolated by hydrodistillation. The resulting oil was colorless with a yield of 1.6% (wt / wt) on a dry-weight basis. Thirty-one components representing 85.3% of the oil were identified by GC-MS (Table 3). The major components were terpinen-4-ol (30.0%), γ-terpinene (11.3%), and *trans*-sabinene hydrate (10.8%). The chemical profile of the oil displayed that oxygenated monoterpenes and monoterpene hydrocarbons were the major groups of compounds and sesquiterpene hydrocarbons and oxygenated sesquiterpenes were minor constituents.

The major constituents of the isolated *M. hortensis* oil were similar to previous reports on the chemistry of this oil isolated from plants growing in Egypt or other countries (Misharina *et al.*, 2003; El-Ghorab *et al.*, 2004); however, the concentrations of the major compounds were different. These changes in essential oil compositions might arise from several environmental (climatical, seasonal, geographical) and genetic differences (Ravid *et al.*, 1992; Perry *et al.*, 1999).

Table 3: Chemical composition of *Majorana hortensis* essential oil.

Compound	RT (min)	Percentage	Compound	RT (min)	Percentage
α-Thujene	9.48	1.08	*trans*-Sabinene hydrate acetate	20.84	1.11
Sabinene	11.04	3.87	Thymol	21.00	0.14
α-Myrecene	11.58	0.98	δ-Elemene	21.76	0.13
α-Phellandrene	12.13	0.98	Neryl acetate	22.44	0.19
α-Terpinene	12.55	6.77	Geranyl acetate	22.97	0.26
p-Cymene	12.80	2.61	β-Caryophyllene	24.13	1.72
γ-Terpinene	12.96	11.34	α-*trans*-Bergamotene	24.38	0.12
cis-Sabinene hydrate	14.29	2.70	α-Humulene	24.99	0.15
Terpinolene	14.70	2.81	Bicyclogermacrene	25.99	0.74
trans-Sabinene hydrate	15.37	10.81	Spathulenol	27.96	0.78
cis-P-Menthen-1-ol	16.09	1.87	Veridiflorol	28.38	0.19
1-Terpineol	16.59	0.54	α-Cadinol	29.43	0.16
Terpinen-4-ol	18.10	29.96	Globulol	33.82	0.11
trans-Piperitol	18.62	0.52	Monoterpene hydrocarbons		30.40
Nerol	18.92	0.20	Oxygenated monoterpenes		48.60
Linalyl acetate	19.57	2.13	Sesquiterpene hydrocarbons		2.90
Piperitone	19.76	0.11	Oxygenated sesquiterpenes		1.20
L-Bornyl acetate	20.50	0.17	Total identified		85.25

Isolation and structure elucidation of the major compounds of *M. hortensis* oil: The two major components of *M. hortensis* oil were isolated by the successive fractionation of the oil on silica gel columns by using acetone/hexane solvent systems. These two compounds were characterized based on ^{1}H and ^{13}C-nuclear magnetic resonance (NMR) and mass spectrometric (MS) data, as terpinen-4-ol and γ-terpinene (Figure1).

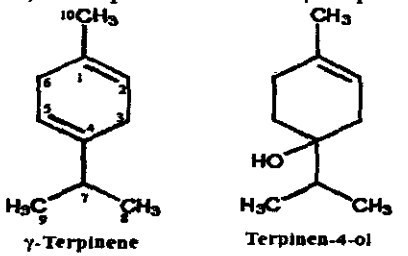

Figure1. Chemical structure of the major constituents of *Majorana hortensis* essential oil.

Insecticidal activity of *M. hortensis* oil and the isolated compounds against *S. littoralis* and *A. fabae*: The insecticidal effects of *M. hortensis* oil and the two isolated compounds (γ-terpinene and terpinen-4-ol) as well as profenofos, a reference insecticide for *S. littoralis*, and methomyl, a reference insecticide for *A. fabae*, are shown in (Table 4). In the case of *S. littoralis*, the essential oil of *M. hortensis* showed a strong insecticidal activity. The LD_{50} value of this oil was 2.48 µg/larva. The oil was more potent than the two isolated compounds (γ-terpinene and terpinen-4-ol). In the meantime, γ-terpinene was more toxic than terpinen-4-ol as their LD_{50} values were 11.86 and16.20 µg/larva, respectively. Despite their strong insecticidal activity as natural products, the oil of *M. hortensis* and its two isolated major compounds (γ-terpinene and terpinen-4-ol) were less effective than profenofos against the larvae *S. littoralis*. Similarly, when tested against *A. fabae*, the *M. hortensis* oil showed higher toxicity than γ-terpinene and terpinen-4-ol. Moreover, γ-terpinene was more effective than terpinen-4-ol. Methomyl was more toxic than the oil, γ-terpinene and terpinen-4-ol.

Table 4: Insecticidal activity of *Majorana hortensis* essential oil, γ-terpinene and terpinen-4-ol against *Spodoptera littoralis* and *Aphis fabae*.

Insect	Compound	LD_{50} (µg/larva) or LC_{50} (g/l)[1]	95% confidence limits	Slope
S. littoralis	Essential oil	2.48 µg/larva	2.24–2.70	3.36 ± 0.32
(Topical	γ-Terpinene	11.86 µg/larva	9.39-13.93	2.40 ± 0.31
application)	Terpinen-4-ol	16.20 µg/larva	13.79-18.42	2.46 ± 0.30
	Profenofos	0.38 µg/larva	0.34-0.44	2.09 ± 0.20
A. fabae	Essential oil	1.86 g/l	1.71-2.00	4.80 ± 0.49
(Rapid dipping)	γ-Terpinene	12.24 g/l	3.15-16.17	4.02 ± 0.48
	Terpinen-4-ol	14.86 g/l	13.43-16.21	3.43 ± 0.45
	Methomyl	0.31 g/l	0.28-0.34	3.50 ± 0.38

The insecticidal activity of *M. hortensis* oil, γ-terpinene and terpinen-4-ol against *S. littoralis* and *A. fabae* has not been reported. Nevertheless, the insecticidal activity of this oil and its major constituents against other economic insects has been studied. For examples, *M. hortensis* oil possessed toxic effect against the larvae and eggs of *Aedes aegypti* (Saleh *et al.*, 1983) and contact toxicity against *Sitophilus oryzae* and *Tribolium castaneum* (Mohamed and Abdelgaleil, 2008). γ-Terpinene and terpinen-4-ol exhibited insecticidal activity against *Leptinotarsa decemlineata* (Kordali *et al.*,

2007). In addition, terpinen-4-ol showed toxic effects on *Sitophilus granarius* and *Sitophilus oryzae* (Lee *et al.*, 2001; Kordali *et al.*, 2006). Moreover, γ-terpinene showed larvicidal activity against *Aedes aegypti* and *Aedes albopictus* (Cheng *et al.*, 2008).

It seems that botanical sources of pesticides will play an important role in future crop protection strategies in response to insect resistance, public health, and environmental concern (Rice *et al.*, 1998). Therefore, the remarkable insecticidal activity of the different extracts and essential oil of *M. hortensis* reported in the present study could be considered as a starting step for use the products of this plant in pest management of *S. littoralis* and *A. fabae*. From an industrial perspective, botanical compounds subject to stringent and robust tests to provide reliable results on a given spectrum of pests, in a range of agronomic and climatic conditions (Mendki *et al.*, 2001). As such, the products of *M. hortensis* require subjection to a range of bioassay procedures, field trials, and toxicity studies before use as pest control agents.

REFERENCES

Abbassy M. A.; A. H. Belal and R. Y. Rabie (2002). Lupin as a source of natural safe insecticides. J. Agric. Environ. Sci. Alex. Univ.1: 51-59.

Abbassy, M. A.; S. A. M. Abdelgaleil and R. Y. Rabie, (2009). Insecticidal and synergistic effects of *Majorana hortensis* essential oil and some of its major constituents. Entomologia Experimentalis et Applicata 131: 225-232.

Abbassy, M. A.; O. A. El-Gougary; Sh. El-Hamody and M. Abu Sholo (1998). Insecticidal, acaricidal and synergistic effects of soosan, *Pancratium maritimum* extracts and constituents. J. Egyp. Soc. Parasitol. 28: 197-198.

Abdelgaleil, S. A. M. (1995). Formulation and efficacy of certain toxicants: Insecticidal efficacy and formulation of certain plant extracts. M. Sc. Thesis, Faculty of Agriculture, Alexandria University, Egypt.

Abdelgaleil, S. A. M.; A. F. El-Aswad and M. Nakatani (2002). Molluscicidal and anti-feedant activities of diterpenes from *Euphorbia paralias* L. Pest Manag. Sci. 58: 479– 482.

Abdel-Rahman, H. and R.N. Al-Mozini (2007). Antifeedant and toxic activity of some plant extracts against the larvae of *Spodoptera littoralis* (Lepidoptera: Noctuidae). Pakistan J. Biochem. Sci.10: 4467-4472.

Abdel-Rasoul, M. A. (2006). Isolation, identification and evaluation the toxicity of biologically active secondary metabolites as pesticides. Ph. D. Thesis, Faculty of Agriculture (Damanhour), Alexandria University, Egypt.

Ahmed, A. A. I.; M. A. Gesraha and C. P. W. Zebitz (2007). Bioactivity of two neem products on *Aphis fabae*. J. Appl. Sci. Res. 3: 392–398.

Batanouny, K. H. (1999). Wild Medicinal Plants in Egypt. An Inventory to Support Conservation and Sustainable Use. The Palm Press, Zamalek, Cairo, Egypt.

Burdock, G. A. (1995). Fenaroli's Handbook of Flavor Ingredients. Vol. I: Natural Flavors. CRC Press, Boca Raton, FL, USA.

Cheng, S. S.; M.-T. Chua; E. H. Chang; C.-G. Huang; W-J. Chen and S.-T. Chang (2008) Variations in insecticidal activity and chemical compositions of leaf essential oils from Cryptomeria japonica at different ages. Bioresour. Technol. 100: 465–470.

Chevallier A. (1996). The Encyclopedia of Medicinal Plants. Dorling Kindersley, London, UK.

Eldefrawi, M. E.; A. Toppozada; N. Mansour and M. Zeid (1964). Toxicological studies on the Egyptian cotton leafworm *Prodenia litura* L. susceptibility of different larval instars of *Prodenia* to insecticides. J. Econ. Entomol. 57: 591–593.

El-Doksh, H. A.; A. M. El-Shazly; A. H. El-Sebae; M. A. Saleh and M. Kady (1984). Plant extracts as feeding deterrents and growth retardants for larvae of the cotton leafworm, *Spodoptera littoralis* (Boisd.). J. Agric. Res. Tanta Univ. 10: 1456-1468.

Elgamal, M. H. A.; K. H. Shaker; K. Pollmann and K. Seifert (1995). Triterpenoid saponins from *Zygophyllum* species. Phytochemistry 40: 1233-1236.

El-Gayar, F. H.; W. M. Watson and M. A. Abbassy (1979). Insecticidal activity of alkaloids isolated from *Datura stromonium* against cotton leafworm, *Spodoptera littoralis* (Boisd.) Proc. 3rd Pesticide Conf. Tanta Univ.1:215-219.

El-Ghorab, A. H.: A. F. Mansour and K. F. El-massry (2004). Effect of extraction methods on the chemical composition and antioxidant activity of Egyptian marjoram (*Majorana hortensis* Moench). Flav. Frag. J. 19: 54–61.

Finney, D. J. (1971). Probit Analysis, 3rd edn. Cambridge University Press, Cambridge, UK.

Guenther, E. (1952) The Essential Oils. Vol. 4: Individual essential oils of the plant families Rutaceae and Labiatae (ed. By Van Nostrand Co.), p. 81. New York, USA.

Kordali, S.; I. Aslan; O. Calmasur and A. Cakir (2006). Toxicity of essential oils isolated from three Artemisia species and some of their major components to granary weevil, *Sitophilus granarius* (L.) (Coleoptera: Curculionidae). Ind. Crops Prod. 23: 162–170.

Kordali, S.; M. Kesdek and A. Cakir (2007). Toxicity of monoterpenes against larvae and adults of Colorado potato beetle, *Leptinotarsa decemlineata* Say (Coleoptera: Chrysomelidae). Ind. Crops Prod. 26: 278–297.

Lee, B. H., W. S. Choi; S. E. Lee and B. S. Park (2001). Fumigant toxicity of essential oils and their constituent compounds towards the rice weevil, *Sitophilus oryzae* (L.). Crop Protect. 20: 317–320.

Mendki, P. S.; V. L. Maheshwari; R. M. Kothari and B. S. Gowda (2001). Botanical pesticides: emerging trends, advantages and limitations. Physiol. Mol. Biol. Plants 7: 107-115.

Misharina, T. A.; A. N. Polshkov; E. L. Ruchkina and I. B. Medvedeva (2003). Changes in the composition of the essential oil of marjoram during storage. Appl. Biochem. Microbiol. 39: 311–316.

Mohamed, M. I. E. and S. A. M. Abdelgaleil (2008). Chemical composition and insecticidal potential the essential oils from Egyptian plants against *Sitophilus oryzae* (L.) and *Tribolium castaneum* (Herbst). Appl. Entomol. Zool. 43 (4): 599–607.

Norman, P. A. and R. A. Sutton (1967). Host plants for melin aphid. J. Econ. Entomol. 60: 1205–1207.

Novak, J.; C. Bitsch; J. Langbehn; F. Pank; M. Skoula; Y. Gotsiou and C. M. Franz (2000). Ratios of cis- and trans-sabinene hydrate in *Origanum majorana* L. and *Origanum microphyllum* (Bentham) Vogel. Biochem. System. Ecol. 28: 697–704.

Pavela, R. (2004). Insecticidal activity of certain medicinal plants. Fitoterapia 75: 745– 749.

Pavela, R. and T. Chermenskaya (2004). Potential insecticidal activity of extracts from 18 species of medicinal plants on larvae of *Spodoptera littoralis*. Plant Protect. Sci. 40: 145–150.

Perry, N. B.; R. E. Anderson; N. J. Brennan; M. H. Douglas; A. J. Heaney; A. J. McGrimpsey and B. M. Smallfield (1999). Essential oil from Dalmation sage (*Salvia officinalis* L.), variations among individuals, plant parts, seasons and sites. J. Agric. Food Chem. 47: 2048–2054.

Prakash A.and J. Rao (2000). Botanical Pesticides in Agriculture. CRC Press, Boca Raton, Florida.

Ravid, U.; E. Putievsky; I. Katzir; D. Carmali; A. Eshel and H. B. Schenk (1992). The essential oil of *Artemisia judaica* L. chemotypes. Flav. Frag. J. 7: 69-72.

Rice, M. J., M. Legg, and K. A. Powell (1998). Natural products in agriculture - a view from the industry. Pestic. Sci. 52: 184-188.

Sadek, M. M. (2003). Antifeedant and toxic activity of *Adhatoda vasica* leaf extract against *Spodoptera littoralis* (Lep., Noctuidae) J. Appl. Entmol. 127: 396–404.

Saleh R. S.; M. S. Saleh and I. A. Gaboub (1983). Management of *Aedes aegypti* by some plant volatile extracts. Proceedings of the International Conference on Environmental Hazards of Agrochemicals in Developing Counties (ed. by A. H. El-Sebae), pp. 1143–1152. Alexandria University, Alexandria, Egypt.

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الخصائص الإبادية الحشرية للمستخلصات النباتية والمونوتربينات على العمر اليرقى الرابع لدودة ورق القطن والحشرات الكاملة لمن الفول

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أختبرت سمية مستخلصات ثلاث نباتات مصرية هى *Zygophyllum coccineum* و *Majorana hortensis* و *Mentha microphylla* على العمر اليرقى الرابع لدودة ورق القطن والحشرات الكاملة لمن الفول. أظهرت جميع المستخلصات المختبرة سمية حشرية ضد الحشرتين على الرغم من أن درجة السمية أعتمدت على نوع المستخلص ونوع النبات و الحشرة المختبرة. مستخلص البترولیم إيثر لنبات *Majorana hortensis* كان أعلى المستخلصات سمية على يرقات دودة ورق القطن حيث كانت قيمة LD_{50} له هى 2.76 مجم/يرقة يليه مستخلص الكلوروفورم لنفس النبات بقيمة LD_{50} تساوى 3.36 مجم/يرقة. مستخلص البترولیم إيثر والإيثانول لنبات *Zygophyllum coccineum* كانا أقل المستخلصات فاعلية ضد دودة ورق القطن. أما فى حالة من الفول فكل من مستخلصات البترولیم إيثر والكلوروفورم لنبات *Majorana hortensis* البترولیم إيثر والإيثانول لنبات *Mentha microphylla* أظهرت سمية عالية حيث كانت قيم LC_{50} 2.21 و 2.44 و 2.62 و 2.25 جم/لتر على الترتيب. الزيت الطيار لنبات *Majorana hortensis* تم إستخلاصة بالتقطير المائي وتم تحليل مكوناتة بواسطة جهاز كروماتوجرافى الغاز/مطياف الكتلة. أتضح أن مركبات terpinen-4-ol (30.0%) و γ-terpinene (11.3%) و *trans*-sabinene hydrate (10.8%) كانت هى المركبات الرئيسية فى هذا الزيت. وقد أظهر الزيت أظهر سمية عالية على دودة ورق القطن ومن الفول. تم عزل مركبى terpinen-4-ol و γ-terpinene بإستخدام الفصل الكروماتوجرافى المتكرر على أعمدة السليكا جل. وقد تم التعرف على التركيب الكيماوى لهذين المركبين بإستخدام الرنين النووى المغناطيسى و مطياف الكتلة. مركب γ-terpinene كان أكثر سمية من مركب terpinen-4-ol على كل من دودة ورق القطن ومن الفول. المركبين المعزولين كانا أقل سمية من الزيت الخام ضد الحشرتين المختبرتين.