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Chemical composition and fumigant toxicity of three citrus essential oils against eggs, larvae and adults of *Callosobruchus maculatus* (Col.: Bruchidae)

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Abstract

The fumigant toxicity of volatile fractions of peel essential oils of the Rutaceae species of *Citrus reticulata* Blanco, *C. limon* L. and *C. aurantium* L. was studied against eggs, larvae and adults of *Callosobruchus maculatus* (F.) at 27 ± 1 ºC and 65 ± 5% RH in darkness. The oils were extracted from the fruit peels using water steam distillation. The essential oils were characterized by a combination of GC and GC/MS analyses. Limonene was the major constituent of the three essential oils. The effect of different concentrations of the essential oil vapors on egg hatchability as well as larval and adult mortality was found to be significant. *Citrus reticulata* and *C. aurantium* oils were more toxic on egg hatchability than *C. limon* extract and caused higher mortality on larvae as well. There was no significant difference between essential oils in terms of adult mortality. The adult beetles were also exposed to the concentrations of 18.5, 37, 55.5 and 74 µl/l air. At the highest concentration (74 µl/l air), *C. aurantium* oil caused 100% mortality after a 6 h exposure, but the oils from *C. reticulata* and *C. limon* caused 38% and 62% mortality, at the identical exposure time, respectively. The results suggest that citrus peel oils can be effectively used as botanical fumigants against various life stages of *Ca. maculatus*.

Key words: fumigant toxicity, essential oil, *Citrus reticulata*, *Citrus limon*, *Citrus aurantium*, *Callosobruchus maculatus*

چکیده

ترکیب شیمیایی و سمیت تدخینی سه اسانس مرکبات روی تخم، لارو و حشره کامل سوسک چهار نقطه‌ای حبوبات، *Callosobruchus maculatus* (Col.: Bruchidae)

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سمیت تنفسی اسانس‌های فرار پوست نارنگی، *Citrus reticulata* Blanco (Rutaceae)، لیمو، *C. limon* L.، و نارنج، *C. aurantium* L.، روی تخم، لارو و حشرات کامل سوسک چهارنقطه‌ای حبوبات، *Callosobruchus maculatus* (F.)، در دمای ۱ ± ۲۷ درجه سلسیوس و رطوبت نسبی ۵ ± ۶۰ درصد در تاریکی مورد بررسی قرار گرفت. اسانس‌ها به روش تقطیر با آب و از پوست میوه‌ها استخراج شدند و با آنالیز GC و GC/MS مورد شناسایی قرار گرفتند. لیمونن (limonene) به‌عنوان مهم‌ترین ترکیب تشکیل‌دهنده اسانس‌ها مد نظر قرار گرفت. اثر غلظت‌های متفاوت بخارات اسانس روی تفریخ تخم و مرگ‌ومیر لارو و حشره کامل معنی‌دار بود .اسانس‌های نارنگی و نارنج نسبت به اسانس لیمو روی میزان تفریخ تخم اثرات سمی‌تری نشان دادند و بیش‌ترین مرگ‌ومیر را نیز روی لاروها ایجاد نمودند. در مورد مرگ‌ومیر حشرات کامل، هیچ اختلاف معنی‌داری بین اسانس‌ها مشاهده نشد. حشرات کامل در معرض غلظت‌های ۱۸/۵، ۳۷، ۵۵/۵ و ۷۴ میکرولیتر بر لیتر هوا قرار گرفتند. اسانس نارنج در بالاترین غلظت (۷۴ میکرولیتر بر لیتر هوا) طی ۶ ساعت، مرگ‌ومیر ۱۰۰ درصد از حشرات را رقم زد. در مقابل اسانس نارنگی و لیمو در زمان مشابه به‌ترتیب موجب مرگ و میر ۳۸ و ۶۲ درصد از حشرات شد. این نتایج پیشنهاد می‌کند که اسانس پوست مرکبات می‌تواند به‌عنوان سموم تدخینی گیاهی در برابر مراحل مختلف سوسک چهارنقطه‌ای حبوبات مورد استفاده قرار گیرد.

واژگان کلیدی: سمیت تدخینی، اسانس، *Citrus reticulata*، *Citrus limon*، *Citrus aurantium*، *Callosobruchus maculatus*

Introduction

The use of fumigants and synthetic insecticides has been a predominant control strategy against insect pest infestation in storage, however their excessive use has led to the development of pest strains resistant to insecticides. In many storage systems, methyl bromide and phosphine are the most economical fumigants for the management of stored-grain insect pests. EPA (2001) proposed a complete halt in the production of methyl bromide by 2005 because of its ozone depletion potential. A demand for safe insecticides remains due to concerns about insecticide residues on grain and a risk of environmental contamination (Subramanyam & Hagstrum, 1995) and has stimulated research into the insecticidal properties of plant natural products. A large number of spices and herbs have insecticidal effects (Tripathi *et al*., 1999), especially in the form of essential oils (Shaaya *et al*., 1991). Essential oils and their constituents are known to be a potent source for botanical pesticides. They are less toxic to warm-blooded animals than other animals, including stored-grain insect pests (Regnault-Roger *et al*., 1993; Shaaya

et al., 1997). Rajapakse (1990) demonstrated that powdered bitter orange peels reduced approximately 45% hatchability of *Callosobruchus maculatus* (F.). In another study, two percent of *Citrus limon* L. leaf powder (W/W) admixed with wheat, caused a 55% reduction of damage by *Trogoderma granarium* Ev. larvae (Jood *et al*., 1993). Other studies assessed the fumigant toxicity of plant derivatives on adults and to lesser extent larvae (Weaver *et al*., 1994; Don-Pedro, 1996b; Negahban *et al*., 2007; Sahaf & Moharramipour, 2008), however little consideration has been given to the egg stage (Ho *et al*., 1997; Huang *et al*., 1997; Tunc *et al*., 2000). The present study was conducted to investigate ovicidal, larvicidal and adultocidal activities of essential oil extractions of *Citrus reticulata* Blanco, *C. aurantium* L. and *C. limon* against *Ca. maculatus*.

Materials and methods

Insect rearing

The specimens of *Ca. maculatus* were reared on mung bean pulses. The cultures were maintained in a dark growth chamber at 27 ± 1 ºC and 65 ± 5% RH. Adult insects, up to three days old, were used for fumigation toxicity tests. All experimental procedures were identical to the environmental condition of the culture.

Plant materials

Fruits of *C. reticulata* (Klemantin cultivar), *C. limon* (Lisbone cultivar) and *C. aurantium* were collected at the ripening stage in the experimental gardens of the Citrus Experimental Institute in the Caspian coast city of Ramsar in December, 2008.

Extraction of essential oils

The essential oils were extracted from fresh rind tissue (albedo and flavedo) by water steam distillation using a Clevenger apparatus until there was no significant increase in the volume of the oil. The essential oils were dried over anhydrous sodium sulfate and stored in glass tubes at +4 ºC in refrigerator. The oils of *C. reticulata*, *C. limon* and *C. aurantium* were 1.71%, 2.38% and 1.46% w/w respectively on the fresh weight basis.

Analysis of essential oils

Gas chromatographic analysis was performed with a Shimadzu GC-9A (Shimadzu, Kyoto, Japan) with helium as a carrier gas with a linear velocity of 30 cm/s on DB-5 column (30 m 25 mm i.d., 0.25 µm film thickness). The oven was programmed to rise to 60 ºC (3 min) isotherm, and then to 210 ºC at a rate of 3 ºC/min. Injector and detector temperatures were 300 ºC and 270 ºC, respectively. The GC/MS analysis was carried out on a Varian 3400 equipped with a DB-5 column with the same characteristics as the one used in GC. The transfer line temperature was 260 ºC. The ionization energy was 70eV with a scan time of 1 s and mass range of 40-300 amu. Identification of the chemical constituents was based on the composition of their relative retention times and mass spectra, either with known compounds or published spectra.

Collection of eggs and larvae

Susceptibility of 1, 4 and 6 day-old eggs to citrus essential oils was tested. To obtain the eggs, 200 adults were introduced to a culture containing of 300 g mung bean to oviposit for one day. Thereafter, all adults were removed from the culture and 1, 4 and 6 day-old eggs collected in the subsequent days. Identical condition (27 ± 1 ºC and 65 ± 5% RH) was provided for the culture and experimental specimens. The larvae of *Ca. maculatus* were 1 day old and 4 and 8 days old.

Fumigant toxicity - Bioassay with immature stages of *Ca. maculatus*

In order to determine the ovicidal and larvicidal activities of citrus essential oil vapors, screw-capped glass jars of 27 ml volume were used as exposure chambers. Essential oil was applied on 2 cm diameter round filter papers inside the caps, using a micro-pipette. The caps were walled up, using laboratory film (parafilm). After dose-setting experiments, final doses

with exposure time of 24 h were used. The mortality for eggs was recorded and the eggs were taken out of the jars and kept in clean glass Petri-dishes. The final mortality was determined after egg hatchability in control glass jars. Unhatched eggs were considered dead and excluded. A stereomicroscope was used to examine the eggs and larvae. Larval mortality count was made possible by splitting the seeds with a fine scalpel. All experimental procedures were carried out at 27 ± 1 ºC and 65 ± 5% RH. Each test was replicated five times.

Fumigant toxicity - Bioassay with adults of *Ca. maculatus*

To assess LC_{50} for adult insects, different concentrations were used to evaluate the mortality of insects after a preliminary dose-setting experiment. Concentrations for *C. reticulata*, *C. limon* and *C. aurantium* oils were from 6.45 to 10.48 µl/l air, 4.43 to 9.27 µl/l air and 7.14 to 17.85 µl/l air, respevtively. Control insects were kept under the same condition without any essential oil. The number of dead and live insects in each bottle was counted 24 h after initial exposure to the essential oil. For both immature and adult stages, probit analysis (Finney, 1971) was used to estimate LC_{50} values with their confidence limits by SAS 6.12 (SAS Institute, 1997). The relative potency was indicative of significant differences between LC_{50}s using SPSS version 16 (Robertson *et al.*, 2007).

To determine the fumigant toxicity of essential oils, the impregnated filter papers (2 cm diameter) were attached inside the screw caps of the glass vials (volume 27 ml) containing 10 (1 day to 3 days old) adult insects. Each concentration and control was replicated five times for each day of the experiment. Mortality was determined separately after 3, 6, 9, 12, 15, 18, 21 and 24 h of their exposure. Motionless insects were presumed dead. There was no dead insect in controls. An experiment was designed to determine median effective time for the mortality of 50% of insects (LT_{50} values) at 18.5, 37, 55.5 and 74 µl/l air of the oil. We followed the method of Finney (1971) to analyze time-mortality data for each experiment, setting the time as explanatory variable to derive estimated hours for 50% mortality (LT_{50}). In this experiment, mortality in each test group was recorded only once. Data resulted from time response experiment was analyzed by separately modelling time trends for each dose. Time as an independent sampling design was analyzed by fitting probit lines to mortality data for a fixed dose over time. Probit analysis was established by computer program written in SPSS version 16.

Results

Chemical constituents of essential oil

The results of the chemical analysis are presented in table 1. Chemical analysis of the essential oil of *C. reticulata* and *C. aurantium* revealed 16 components in which limonene, linalool and α-pinene were the major constituents. Essential oil of *C. limon* contained 17 compounds, including limonene (73.25%), β-pinene (8.44%), γ-terpinene (6.21%) and geraniol (2.53%).

Fumigant toxicity

Based on the LC_{50} values, the eggs were significantly more susceptible to *C. reticulata* than both *C. limon* and *C. aurantium* essential oils. No significant differences were observed between *C. limon* and *C. aurantium*. In all cases, the young eggs were more tolerant to essential oil vapors than the older ones. Apart from the inhibition of hatching, the exposure of eggs to essential oil vapors increased post-embryonic mortality of the larvae. In terms of LC_{50}, *Ca. maculatus* larvae were more sensitive to *C. aurantium* and *C. reticulata* essential oils. LC_{50} values for 8-day-old larvae of *C. reticulata*, *C. limon* and *C. aurantium* oils were 143.09, 169.78 and 110.13 µl/l air, respectively. The effect of the oil on the different larval stages inside the seed was dependent on the age of the larvae as 1-day-old larvae were found to be more susceptible than 4 and 8-day-old individuals. The mortality of different life stages of *Ca. maculatus* after treatment with citrus essential oils appear in tables 2, 3 and 4. Experiments were conducted to determine

whether the insecticidal activity of *C. reticulata*, *C. limon* and *C. aurantium* oils was related to fumigant action. In all cases, the mortality was proportional to the concentration of essential oils and exposure times. The oil extracted from *C. aurantium* showed strong fumigant activity against adults at different concentrations and exposure times (fig. 1), although based on relative potency, there was no significant difference between essential oils against adults. All concentrations of *C. aurantium* oil caused at least 90% mortality after a 24 h exposure. At the highest concentration (74 µl/l air), *C. aurantium* oil caused 64% mortality with a 3 h exposure and 100% mortality after a 6 h exposure. By contrast, only 38% and 62% mortality was achived by *C. reticulata* and *C. limon* essential oils respectively during a 6 h exposure. *Citrus limon* oil at 74 µl/l air caused 100% mortality in *Ca. maculatus* within a 15 h, 18 h, 21 h and 24 h exposure (fig. 1). Small quantity of Chi-square in our results suggests that our probit model is fitted. Also differences in slopes, in different treatments show significant differences with relative potency.

LT_{50} values was disproportionate to the concentraton of essential oils. LT_{50} value of *C. reticulata* decreased from 23.66 h (95% lower and upper confidence limits (CL) = 19.13-34.90 h) at concentration 18.5 µl/l air to 8.24 h (95% CL = 3.99-11.52 h) at concentration 74 µl/l air. At the same concentration, the LT_{50} of *C. limon* decreased from 17.50 h (95% CL = 15.49-20.37 h) to 4.92 h (95% CL = 0.50-7.78 h) and LT_{50} value of *C. aurantium* descended from 15.44 h (95% CL = 11.30-20.37 h) to 2.61 h (95% CL = 0.50-3.25 h), respectively. However, based on LT_{50} values, there was no significant difference between the essential oils of *C. aurantium* and *C. limon* at 55.5 µl/l air.

Table 1. Major chemical composition of the essential oils extracted from *Citrus reticulata*, *C. limon* and *C. aurantium.*

Compounds	Retention Index	Chemical compounds (%)		
		Citrus reticulata	*Citrus limon*	*Citrus aurantium*
α-pinene	935	-	-	4.70
β-pinene	976	-	8.44	-
Limonene	1027	83.8	73.25	87.61
γ-terpinene	1059	-	6.21	-
Linalool	1096	4.3	-	-
n-decanal	1199	2.2	-	-
Neral	1240	-	2.16	-
Geranial	1271	-	2.53	-
Other compounds	-	9.7	7.41	7.69

Table 2. Fumigant toxicity of *Citrus reticulata*, *C. limon* and *C. aurantium* essential oils against *Callosobruchus maculatus* eggs.

Essential oils	Age of eggs (days old)	LC_{50}[a] (µl/l air)	Slope ± SE	Chi square (χ^2)	Degree of freedom	N	P-value
C. reticulata	1	36.60 (33.83-39.45)	1.73 ± 0.82	0.07	3	200	0.996
C. reticulata	4	25.75 (21.93-30.28)	2.46 ± 0.39	2.10	4	250	0.715
C. reticulata	6	19.91 (15.94-23.97)	2.95 ± 0.32	2.29	4	250	0.681
C. limon	1	54.52 (49.63-60.96)	3.47 ± 0.50	1.14	6	350	0.961
C. limon	4	38.82 (35.34-43.09)	4.40 ± 0.76	0.03	3	200	0.992
C. limon	6	29.28 (25.98-32.98)	4.61 ± 0.60	1.03	3	200	0.792
C. aurantium	1	53.37 (45.68-60.1)	1.47± 1.72	3.02	4	250	0.961
C. aurantium	4	34.83 (30.00-41.58)	2.41 ± 0.42	7.27	5	300	0.992
C. aurantium	6	27.86 (24.22-31.51)	2.95 ± 0.43	5.72	5	300	0.792

a: 95% lower and upper confidence limits are shown in parenthesis.

Table 3. Fumigant toxicity of *Citrus reticulata*, *C. limon* and *C. aurantium* essential oils against *Callosobruchus maculatus* larvae.

Essential oils	Age of larvae (days old)	LC_{50}[a] (µl/l air)	Slope ± SE	Chi square (χ^2)	Degree of freedom	N	P-value
C. reticulata	1	70.33 (65.21-76.44)	5.14 ± 0.75	0.65	4	250	0.952
C. reticulata	4	114.09 (109.12-120.16)	8.80 ± 1.29	0.97	4	250	0.911
C. reticulata	8	143.09 (136.81-151.06)	8.95 ± 1.15	1.07	5	300	0.782
C. limon	1	85.90 (81.10-91.68)	7.27 ± 1.19	0.62	3	200	0.881
C. limon	4	147.21 (141.37-154.43)	4.40 ± 0.76	0.77	5	300	0.971
C. limon	8	169.78 (165.18-174.79)	6.29 ± 0.92	0.26	4	250	0.993
C. aurantium	1	61.43 (56.25-67.07)	4.24 ± 0.56	2.22	5	300	0.881
C. aurantium	4	89.42 (80.97-95.51)	7.38 ± 1.50	6.84	3	200	0.971
C. aurantium	8	110.13 (105.58-114.79)	9.52 ± 1.25	1.66	4	250	0.993

a: 95% lower and upper confidence limits are shown in parenthesis.

Table 4. Fumigant toxicity of *Citrus reticulata*, *C. limon* and *C. aurantium* essential oils against *Callosobruchus maculatus* adults.

Essential oils	Age of adults (days old)	LC_{50}[a] (µl/l air)	Slope ± SE	Chi square (χ^2)	Degree of freedom	N	P-value
C. reticulata	1-3	8.70 (8.30-9.15)	8.06 ± 1.23	0.77	4	250	0.941
C. limon	1-3	7.21 (6.79-7.71)	5.81 ± 0.82	5.04	5	300	0.410
C. aurantium	1-3	6.33 (5.88-6.88)	5.02 ± 0.85	0.61	4	250	0.410

a: 95% lower and upper confidence limits are shown in parenthesis.

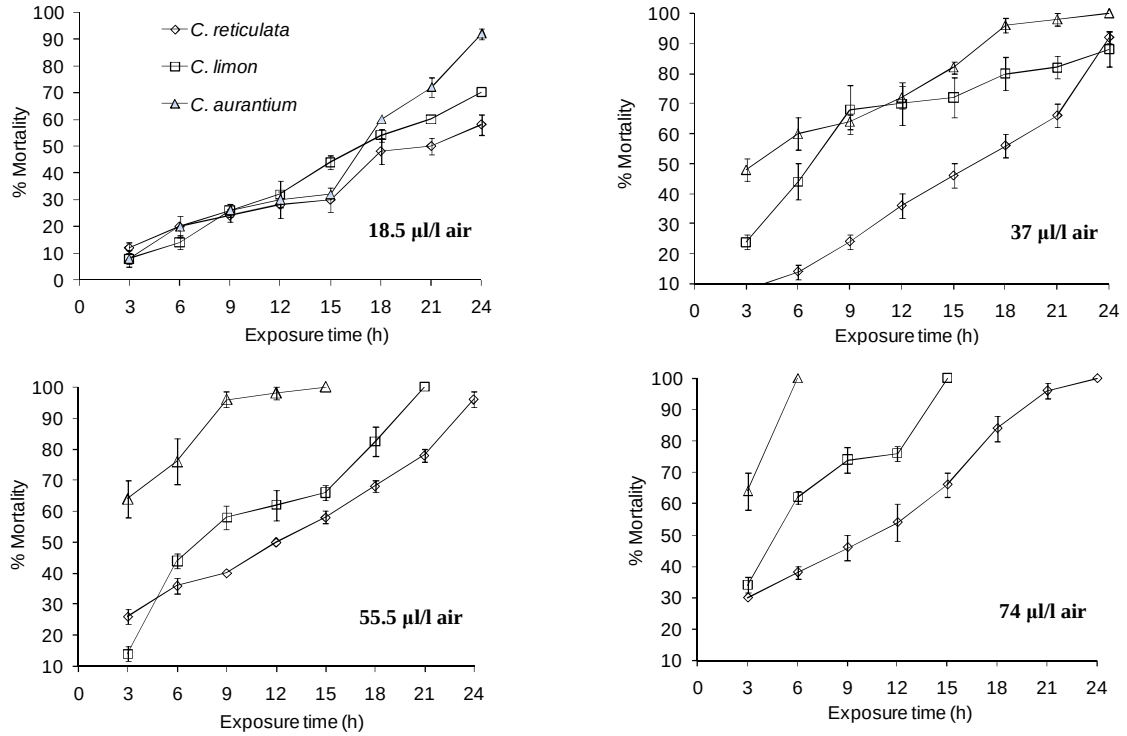

Fig. 1. Percentage mortality of *Callosobruchus maculatus* exposed to various periods of time to essential oils from *Citrus reticulata*, *C. limon* and *C. aurantium*.

Discussion

Over 120 plants and plant products especially essential oils have been shown to have insecticidal activity against stored product pests (Dale, 1996; Isman, 2000). Jacobson (1989) pointed out that the most promising botanical insect control agents are in the families Annonaceae, Asteraceae, Canellaceae, Lamiaceae, Meliaceae and Rutaceae. The family Rutaceae is a large family containing 130 genera in seven subfamilies, with many important fruits and essential oil products. In this experiment, potent toxicity was exhibited by *C. reticulata*, *C. limon* and *C. aurantium* oils against eggs, larvae and adults of *Ca. maculatus*. We found that toxic vapor was able to penetrate the seeds and kill the immature stages inside the beans. The fumigant activity of the essential oils could be largely attributed to limonene as the major component of the oils. Lee *et al.* (2002) tested 24 essential oils for their fumigant toxicity on *Sitophilus oryzae* (L.), *Oryzaephilus surinamensis* L., *Tribolium castaneum* (Herbst), and found that limonene caused a 100% mortality in all species. Karr & Coats (1988) noted that high concentrations of limonene vapors are effectively lethal against German cockroach. Linalool, β-pinene and α-pinene were other constituents of the tested essential oils. Fumigant toxicity of linalool has been demonstrated on *Rhyzoptera dominica* F. (Rozman *et al.*, 2006). α-pinene is reported to be toxic to *Tribolium confusum* Jacquelin Du Val (Ojimelukwe & Alder, 1999). Therefore, the fumigant toxicity of citrus essential oils in the present study could be attributed to these compounds. A common structural feature of terpenoids is their hydrocarbon skeleton, which in turn renders them hydrophobic. Many hydrophobic compounds are associated with protein or enzyme deactivation, where acetylcholinesterase is particularly sensitive. Compounds that inhibit or inactivate acetylcholinesterase, cause acetylcholine to accumulate at synapses of cholinergic sites. In general, the most possible target for essential oil neurotoxicity is the octopaminergic system in insects because they are similar to octopamine neurotransmitter (Enan, 2001).

Eggs of *Ca. maculatus* were significantly susceptible to *C. reticulata* essential oil, likely due to the presence of linalool. Linalool is a terpenoid alcohol that irritates nervous system and causes death in some insects like *R. dominica* and ants (Rozman *et al.*, 2006). The essential oil of *C. aurantium* showed the highest activity against the larvae and adults of *Ca. maculatus.* The insecticidal activity varied with plant species, concentration of the oil and exposure time. In our experiments, essential oils were highly effective when eggs had been exposed to the oils for 24 h. In fact, the more embryonic development advances the greater the susceptibility occurs. Papachristos & Stamopoulos (2002) found that the degree of susceptibility of the eggs of *Acanthoselides obtectus* (Say) to essential oil vapors varied with age. It can be concluded that the ovicidal activity may become apparent when the target system (the nervous system) begins to develop (Smith & Salkeld, 1966; Michaelides & Wright, 1997). Alternatively, changes in the permeability of the chorion and vitelline membrane may occur during embryogenesis and may facilitate the diffusion of vapors into older eggs (Gurusubramanian & Krishna, 1996). Although some researchers have found a negative correlation between age and susceptibility (Risha *et al.*, 1990; Rahman & Schmidt, 1999), others found a positive one (Saxena & Srivastava, 1972). Other factors such as the insect species, essential oil and chemical compounds may be involved. Comparing with the study of Sahaf & Moharramipour (2008), our findings demonstrated higher toxicity of citrus oils than *Carum copticum* C. B. Clarke and *Vitex pseudo-negundo* Hand I. MZT against larvae of *Ca. maculatus.* The results from Shakarami *et al.* (2003, 2004, 2005) show that the essential oils of *Artemisia aucheri* Boiss, *Salvia bracteata* L. and *Nepeta cataria* L. against eggs, larvae and adults of *Ca. maculatus* are less effective than the extractions of *C. reticulata*, *C. limon* and *C. aurantium*. Don-pedro (1996a) attributed the mortality of *Ca.*

maculatus adults on citrus peel-treated grains to the fumigant activity of the vapor released from the peels. He indicated that grains treated by 7 ml oil/kg^{-1} seed caused 100% mortality after 1 h exposure. However, we did not use essential oils admixed with cowpea grains. In addition to insecticidal activity, antimicrobial properties of the citrus oils have been an interesting field in food and cosmetics industries. These studies indicate that citrus essential oil can be a potential protectant by killing various life stages of *Ca. maculatus* through their strong fumigant activity. It is also necessary to study the durability or possible adverse effect of essential oils on the quality of stored products. Further studies are required to improve the efficiency of citrus oils stored-product insect pests. The volatility, poor water solubility, aptitude for oxidation and high sorption of essential oils underscores the limiting factors for any successful application of natural compounds in large scale commodity fumigations. To overcome these restrictions, essential oils can be manufactured as nanoencapsuled formulations to gradually release their active ingredients. Using botanical products in pest management programs are becoming increasingly economical and environment-friendly (Negahban *et al.*, 2013).

References

Dale, M. J. (1996) *A review of plant material used for controlling insect pests of stored products*. 84 pp. Natural Resources Institute, London.

Don-Pedro, K. N. (1996a) Fumigant toxicity is the major route of insecticidal activity of citrus peel essential oils. *Pesticide Science* 46, 71-78.

Don-Pedro, K. N. (1996b) Fumigant toxicity of citrus peel oils against adult and immature stages of storage insect pests. *Pesticide Science* 47, 213-223.

Enan, E. (2001) Insecticidal activity of essential oils: octopaminergic sites of action. Inhibition of acetylcholinesterase by pulegone-1, 2-epoxide. *Pesticide Biochemistry and Physiology* 23, 383-388.

EPA. (2001) Protection of stratospheric ozone: process for exempting quarantine and pre shipment application of methyl bromide. United States Environmental Protection Agency (EPA), 37752-37769.

Finney, D. J. (1971) *Probit analysis.* 3rd ed. 333pp. Cambridge University Press, Cambridge.

Gurusubramanian, G. & Krishna, S. S. (1996) The effects of exposing eggs of four cotton insects pests to volatiles of *Allium sativum* (Liliaceae). *Bulletin of Entomological Research* 86, 29-31.

Ho, S. H., Ma, Y. & Huang, Y. (1997) Anethol, a potential insecticide from *Illicium verum* Hook F., against two stored-product insects. *International Pest Control* 39, 50-51.

Huang, Y., Tan, J. M. W. L., Kini, R. M. & Ho, S. H. (1997) Toxic and antifeedant action of nutmeg oil against *Tribolium castaneum* (Herbst) and *Sitophilus oryzae* Motsch. *Journal of Stored Products Research* 33, 289-298.

Isman, M. B. (2000) Plant essential oils for pest and disease management. *Crop Protection* 19, 603-608.

Jacobson, M. (1989) Botanical pesticide: past, present, and future. pp. 1-10 in Arnason, J. T., Philogene, B. J. R. & Morand, P. (Eds) *Insecticides of plant origin.* 213 pp. American Chemical Society. Washington DC.

Jood, S., Kapoor, A. C. & Singh, R. (1993) Evaluation of some plant products against *Trogoderma granarium* Everts in stored wheat and their effects on nutritional composition and organoleptic characteristics of treated grains. *International Journal of Pest Management* 39, 93-98.

Karr, L. L. & Coats, J. R. (1988) Insecticidal properties of *d*-limonene. *Journal of Pesticide Science*13, 287-290.

Lee, S., Peterson, C. J. & Coats, J. R. (2002) Fumigant toxicity of monoterpenoids to several stored product insects. *Journal of Stored Product Research* 39, 77-85.

Michaelides, P. K. & Wright, D. J. (1997) Activity of soil insecticides on eggs of *Diabrotica undecimpunctata howardi*, effects on embryological development and influence of egg age. *Pesticide Science* 49, 1-8.

Negahban, M., Moharramipour, S. & Sefidkon, F. (2007) Fumigant toxicity of essential oil from *Artemisia sieberi* Besser against three stored-product insects. *Journal of Stored Products Research* 43, 123-128.

Negahban, M., Moharramipour, S., Zandi, M. & Hashemi, S. A. (2013) Efficiency of nanoencapsulated essential oil of *Artemisia sieberi* on nutritional indices of *Plutella xylostella*. *Iranian Journal of Medicinal and Aromatic Plants* 29, 692-708.

Ojimelukwe, P. C. & Alder, C. (1999) Potential of zimtaldehyde, 4- allyl-anisol, linalool, terpineol and other phytochemicals for the control of confused flour Beetle (*Tribolium confusum* J. D. V) (Coleoptera: Tenebrionidae). *Journal of Pesticide Science* 72, 81-86.

Papachristos, D. P. & Stamopoulos, D. C. (2002) Toxicity of vapors of three essential oils to the immature stages of *Acanthoselides obtectus* (Say) (Coleoptera: Bruchidae). *Journal of Stored Product Research* 38, 365-373.

Rahman, M. M. & Schmidt, G. H. (1999) Effects of *Acorus calamus* (L.) (Araceae) essential oil vapors from various origins on *Callosobruchus phaseoli* (Gyllenhal) (Coleoptera: Bruchidae). *Journal of Stored Products Research* 38, 117-128.

Rajapakse, R. H. S. (1990) Effect of five botanicals as protectants of green gram against the pulse beetle *Callosobruchus maculatus*. pp. 85-90 in Fujii, K. (Ed.) *Bruchids and legumes*: *economics, ecology and coevolution*. 407 pp. Kluwer Academic Publishers, The Netherlands.

Regnault-Roger, C., Hamraoui, A., Holeman, M., Theron, E. & Pinel, R. (1993) Insecticidal effect of essential oils from Mediterranean plants upon *Acanthoselides obtectus* (Say) (Coleoptera: Bruchidae), a pest of kidney bean (*Phaseolus vulgaris* L.). *Journal of Chemical Ecology* 14, 1965-1975.

Risha, E. M., El-Nahal, A. K. M. & Schmidt, G. H. (1990) Toxicity of vapors of *Acorus calamus* L. oil to the immature stages of some stored-product Coleoptera. *Journal of Stored Products Research* 26, 133-137.

Robertson, J. L., Savin, N. E., Preisler, H. K. & Russell, R. M. (2007) *Bioassays with arthropods*. 2nd ed. 224 pp. CRC Press Boca Raton, Fl, USA.

Rozman, V., Kalinovic, I. & Korunic, Z. (2006) Toxicity of naturally ocuring compounds of Lamiaceae and Luraceae to three stored-product insects. *Journal of Stored Products Research* 43, 349-355.

Sahaf, B. Z. & Moharramipour, S. (2008) Fumigant toxicity of *Carum copticum* and *Vitex pseudo-negundo* essential oils against eggs, larvae and adults of *Callosobruchus maculatus*. *Journal of Pest Science* 81, 213-220.

SAS Institute (1997) *SAS users guide*. SAS Institute, USA, 135.

Saxena, B. P. & Srivastava, J. B. (1972) Effects of *Acorus calamus* L. oil vapors on *Dysdercus koenigii* F. *Indian Journal of Experimental Biology* 10, 391-393.

Shaaya, E., Kostjukovski, M., Eilberg, J. & Sukprakarn, C. (1997) Plant oil as fumigants and contact insecticides for the control of stored-product insects. *Journal of Stored Product Research* 33, 7-15.

Shaaya, E., Ravid, U., Paster, N., Juven, B., Zisman, U. & Pisarrev, V. (1991) Fumigant toxicity of essential oils against four major stored-product insects. *Journal of Chemical Ecology* 17, 499-504.

Shakarami, J., Kamali, K. & Moharramipour, S. (2005) Fumigant toxicity and repellency effect of essential oil of *Salvia bracteata* on four species of warehouse pests. *Journal of Entomological Society of Iran* 24, 35-50.

Shakarami, J., Kamali, K., Moharramipour, S. & Meshkatalsadat, M. (2003) Fumigant toxicity and repellency of essential oil of *Artemisia aucheri* on four species of stored pest. *Applied Entomology Phytopathology* 71, 61-75.

Shakarami, J., Kamali, K., Moharramipour, S. & Meshkatalsadat, M. (2004) Effects of three plant essential oils on biological activity of *Callosobruchus maculatus* F. (Coleoptera: Bruchidae). *Iranian Journal of Agricultural Science* 35, 965-972.

Smith, E. H. & Salkeld, H. E. (1966) The use and action of ovicides. *Annual Review of Entomology* 11, 331-368.

Subramanyam, Bh. & Hagstrum, D. (1995) Resistance measurement and management. pp. 331-398 in Subramanyam, Bh. & Hagstrum, D. (Eds) *Integrated management of insects in stored products*. 432 pp. Marcel Dekker, New York.

Tripathi, A. K., Prajapati, V., Gupta, R. & Kumar, S. (1999) Herbal material for the insect-pest management in stored grains under tropical conditions. *Journal of Medicinal and Aromatic Plant Science* 21, 408-430.

Tunc, I., Berger, B. M., Erler, F. & Dagli, F. (2000) Ovicidal activity of essential oils from five plants against two stored-product insects. *Journal of Stored Products Research* 36, 161-168.

Weaver, K. D., Dunkel, V. F., Potter, C. R. & Ntezuubanza, L. (1994) Contact and fumigant efficacy of powdered and intact *Ocimum canum* Sims (Lamiales: Lamiaceae) against *Zabrotes subfasciatus* (Boheman) adults (Coleoptera: Bruchidae). *Journal of Stored Products Research* 30, 243-252.

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