Effectiveness of monoterpenes against Sitophilus oryzae (L.) and Tribolium castaneum (Herbst) in stored wheat

Sailan A. A. El-arami^a, Magdy I. E. Mohamed^b, Samir A. M. Abdelgaleil^{c,*}

^aDepartment Plant Protection, Faculty of Agriculture, Sanaa University, Sanaa, Yemen
^bDepartment of Stored Product Pests, Plant Protection Research Institute, Agricultural
Research Center, Sabahia, Alexandria, Egypt

Department of Pesticides Chemistry, Faculty of Agriculture, Alexandria University, 21545-El-Shatby, Alexandria, Egypt

ABSTRACT

A study was conducted to evaluate the efficacy of eight monoterpenes namely (-)-carvone, 1-8-cineole, cuminaldehyde, (L)-fenchone, geraniol, (-)-limonene, (-)-linalool and myrcene against two major stored-grain insects on wheat seeds in the laboratory. Insect species tested were the rice weevil, Sitophilus oryzae (L.), and the rust red flour beetle, Tribolium castaneum (Herbst). Wheat seeds were treated with monoterpenes at 0.5, 1 and 5 mg/g for controlling the two insect species. Out of the eight monoterpenes, (-)-carvone was the most toxic compound against both insects at the three tested application rates after one and two weeks of treatment. Geraniol and cuminaldehyde were among the most effective compounds against both insects. Four of the tested monoterpenes (carvone, cuminaldehyde, geraniol and (-)-linalool) caused complete mortality to the adults of S. oryzae after one week at the rate of 5mg/g, while all of the tested monoterpenes except myrcene and (-)-linalool caused complete mortality of T. castaneum adults at the same application rate. Comparing the toxicity of monoterpenes toward the two insects indicated that, -(-)limonene, (L)-fenchone and cuminaldehyde had approximately the same activity on both insects. On the other hand, myrcene, geraniol, (-)-carvone and 1-8-cineole were more toxic against T. castaneum than S. oryzae, while (-)-linalool was more toxic towards S. oryzae than T. castaneum. All of the tested compounds except for myrcene showed significant reduction in progeny production of S. oryzae after 6 and 12 weeks of treatment compared with control. (-)-Carvone was the most effective in reducing the progeny production at the three tested application rates, followed by geraniol, cuminaldehyde and (-)-linalool. These four monoterpenes provided complete suppression of progeny production at the rate of 5 mg/g after 12 weeks. At the rates of 1 and 5mg/g, the majority of monoterpenes significantly reduced weight loss of treated wheat and complete protection was observed in wheat treated with (-)-carvone, geraniol, cuminaldehyde and (-)-linalool at 5 mg/g. These strong insecticidal activities of monoterpenes particularly (-)-carvone, geraniol, cuminaldehyde and (-)-linalool indicate that these monoterpenes can used for control S. oryzae and T. castaneum in stored wheat.

Keywords: Monoterpenes; insecticidal activity; *Sitophilus oryzae*; *Tribolium castaneum*; stored wheat

INTRODUCTION

Losses of grain in storage due to insects are main components of the insect damage in agricultural production. These losses can exceed those incurred while growing the crop. Insects damage stored grain directly by consuming kernels and reducing the weight and feed value of the infested commodity. Additionally, insects infesting stored grain produce heat and moisture that contribute to mold growth and spoilage (Evans, 1987). Worldwide losses in stored products, caused by insects, have been estimated to be between five and ten percent. Heavier losses occurring in the tropics may reach 30% (Larry, 2000).

Insect control in stored grain at present relies heavily upon the use of fumigants (methyl bromide and phosphine) and protectants (synthetic insecticides). However, the intensive use of these chemicals may led to some environmental and health problems as well as the development of insect resistance (Collins et al., 1993). For this reason, alternative control means are necessary. Plant secondary metabolites may provide potential alternatives to currently used insect control agents. The plant materials have several advantages over traditional pest control agents; such as specificity, biodegradability and low mammalian toxicity (Wink, 1993; Mohan and Fields, 2002).

One of the most promising secondary metabolites in the field of pest control is monoterpenes which are common constituents of plant essential oils. They are biosynthesized from geranyl pyrophosphate, the ubiquitous acyclic C_{10} intermediate of the isoprenoid pathway (Windholz *et al.*, 1983). Monoterpenes can be classified into two major groups: monoterpene hydrocarbons and oxygenated monoterpenes. The later group includes alcohols, aldehydes, ketones, ethers and acids (Templeton 1969).

Monoterpenes have been considered as potential pest control agents. This includes their insecticidal activity (Waliwitiya et al., 2005; Choi et al., 2006; Kordali, et al., 2006; Samarasekera et al., 2008), herbicidal activity (Kohli et al., 1998; Isman 2000; Romagni et al., 2000), repellent activity (Mason, 1990; Watanabe et al., 1993), antifeedant activity (Hough-Goldstein, 1990; Hummelbrunner and Isman, 2001; Argandoña et al., 2002) and development and growth inhibitory activity (Gunderson et al., 1985; Karr and Coats 1992). The wide spectrum of the biological activities of monoterpenes is attributed to their physical-chemical properties such as lipophilicity and low vapour pressure.

Although, the fumigant and contact toxicities of some monoterpenes have been studied on the rice weevil, Sitophilus oryzae (Lee et al. 2001, 2003 and 2004, Kim and Ahn 2001; Park et al., 2003) and the rust red flour beetle, Tribolium castaneum (Rice and Coats, 1994; Partes et al., 1998; Lee et al., 2003, 2004), there were no reported studies on the effectiveness of monoterpenes for the control of both insects on the stored wheat. Therefore, the present study was conducted to investigate the potential use of monoterpenes for the control of these two important stored product insects in stored wheat. The effect of the monoterpenes on the progeny production of S. oryzae and weight loss of wheat seeds was also investigated.

MATERIALS AND METHODS

Monoterpenes: The monoterpenes, (-)-carvone (98%), 1-8-cineole (99%), cuminaldehyde (98%), (L)-fenchone (98%), geraniol (98%), (-)-limonene (96%), (-)-linalool (95%) and myrcene (90%) were purchased from Sigma-Aldrich Chemical Co., Steinheim, Germany. The chemical structures of the tested monoterpenes are shown in (Figure 1).

Insects and commodity: Sitophilus oryzae (L.) (Coleoptera: Curculionidae) adults were taken from culture that was kept on whole hard wheat in the laboratory at 26±1°C and 65±5% R.H. Tribolium castaneum (Herbst) (Coleoptera: Tenebrionidae) adults were taken from a culture kept on wheat flour mixed with yeast (10:1, w/w) under the same conditions. The colonies of both insects were maintained in our laboratory over 10 years without exposure to insecticides. The adults used in the tests were 2 weeks postemergence. Untreated, clean, sterilized and infestation-free Egyptian wheat with 13.6% moisture content was used in the tests for two insect species.

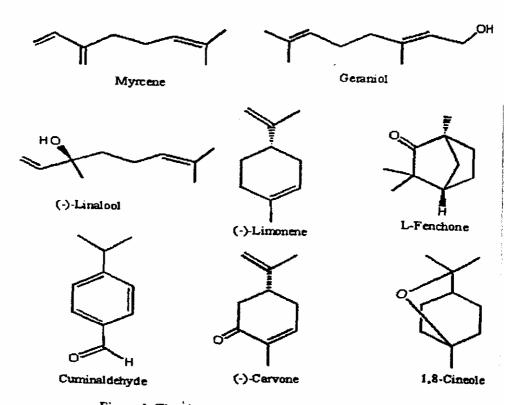


Figure 1. The chemical structures of the tested monoterpenes

Bioassay: Stock solutions of the tested monoterpenes were prepared in acetone. Fifty grams of wheat were placed in 300 ml glass jars. Wheat in glass jars were treated with 1 ml of the stock solution of the tested monoterpenes. The application rates were 0.5, 1 and 5 mg/g. The jars were shaken manually for 3 minutes to achieve distribution of the compounds throughout the grains. The control grains were treated with acetone only. The jars were left for 30 minutes for complete evaporation of the solvent. Twenty adults of S. oryzae and T. castaneum were separately introduced into each jar. Three replicates were used for each treatment and control. The jars were covered with cheese cloth fastened by rubber bands to prevent escape of insects and to ensure proper ventilation. The jars were kept at $26\pm1^{\circ}\text{C}$ and $65\pm5\%$ R.H. The adult mortality was done at one week from

the initial treatment date at which time all dead insects were counted and removed. After another week, adult mortality was assessed again giving a cumulative 2 weeks mortality assessment, with all remaining adults being removed from the treatment. In the assessment of adult mortality, insects were recorded as dead if they were unable to move after gentle prodding with a fine-haired brush or/and unable to move or walk during a 2 minutes observation period. After the last mortality count, all jars were retained under the same conditions as above. After 6 and 12 weeks later, the number of emerged adults of S. oryzae were counted and expressed as progeny production. The following formula was used to determine the reduction percentage in the number of progeny after 6 and 12 weeks of treatment: $\% = 1 - x/y \times 100$, where x = the number of adults emerging in the treatment; y = the number of adults emerging in the control. The grains were sieved and the powder was discarded. The weight of remaining grains in treatments and control was recorded. The weight loss percentage was calculated from the following formula:

% Weight loss = $Wu - Wi/Wu \times 100$, where Wu = weight of uninfested grain; Wi = weight of infested grain of control and treatment.

Grain germination: In order to assess the effect of tested monoterpenes on the viability of grains, grains germination was tested using 100 randomly picked grains from each jar treated with the higher application rate (5 mg/g). This test was done on six monoterpenes which showed the highest or/and complete protection of the grains. The seeds were placed on a moistened filter paper in glass Petri dishes and the number of germinated grains was recorded after one week.

Statistical analysis: Statistical analyses were conducted using the statistical package SPSS version 12.0 (Statistical Package for Social Sciences, USA). The mortality, progeny and weight loss data were submitted to a one-way analysis of variance (ANOVA). Mean separations were performed by Student-Newman-Keuls (SNK) test and differences at P = 0.05 were considered as significant.

RESULTS AND DISCUSSION

Effect of monoterpenes on mortality of S. oryzae and T. castaneum: The effect of different monoterpenes on the mortality of S. oryzae adults after one and two weeks of treatment is presented in Table 1.

El-arami, S. A. A. et.al.,

Table 1. Effect of monoterpenes against *Sitophilus oryzae* exposed for 1 and 2 weeks on treated wheat.

Monoterpene class	Monoterpene	Application rate (mg/g)	Cumulative mortality (% ± SE)		
Class			1 Week	2 Weeks	
	Control	0	0.0 ± 0.0^{g}	3.3 ± 1.67^{ef}	
Hydrocarbon	(-)-Limonene	0.5	0.0 ± 0.0^{g}	1.7 ± 1.67^{f}	
		1	$10.0.\pm 2.89^{fg}$	13.7 ± 1.67^{def}	
		5	88.3 ± 4.41^{ab}	91.7 ± 1.67^{a}	
	Myrcene	0.5	0.0 ± 0.0^{g}	1.7 ± 1.67^{f}	
		1	0.0 ± 0.0^{g}	1.7 ± 1.67^{f}	
		5	5.0 ± 2.89^{fg}	8.3 ± 1.67^{def}	
Alcohol	Geraniol	0.5	$26.7 \pm 4.41^{\circ}$	51.7 ± 4.41^{b}	
		1	$85.0 \pm 2.89^{\circ}$	93.3 ± 4.41	
		5	100.0 ± 0.0^{a}	100.0 ± 0.0^{a}	
	(-)-Linalool	0.5	8.3 ± 3.34^{fg}	10.0 ± 2.89^{def}	
		1	$46.7 \pm 3.34^{\circ}$	46.7 ± 3.34^{b}	
		5	100.0 ± 0.0^{a}	100.0 ± 0.0^{a}	
Ketone	(-)-Carvone	0.5	36.7 ± 1.67^{d}	$36.7 \pm 1.67^{\circ}$	
		1	$96.7 \pm 3.34^{\circ}$	96.7 ± 3.34^{a}	
		5	100.0 ± 0.0^{a}	100.0 ± 0.0^a	
	(L)-Fenchone	0.5	5.0 ± 0.0^{fg}	11.7 ±1.67 ^{def}	
		1	5.0 ± 2.89^{fg}	11.7 ± 1.67^{def}	
		5	91.7 ± 4.41^{ab}	91.7 ± 4.41^{6}	
Aldehyde	Cuminaldehyde	0.5	15.0 ± 2.89^{f}	16.7 ± 3.34^{d}	
		1	88.3 ± 3.34^{ab}	90.0 ± 2.89^{a}	
		5	100.0 ± 0.0^{1}	100.0 ± 0.0^a	
Oxide	1-8-Cineole	0.5	0.0 ± 0.0^{g}	3.3 ± 1.67^{ef}	
		1	6.7 ± 1.67^{fg}	15.0 ± 2.89^{de}	
		5	$50.0 \pm 5.0^{\circ}$	51.7 ± 4.41^{b}	

Means in a column with same letters are not-significant (p < 0.05) compared to the control.

The results showed that there were significant effects between monoterpenes and among the concentrations of each monoterpene on the percentage adult mortality. At the application rate of 0.5 mg/g, (-)-carvone showed the highest mortality (36.7%) followed by geraniol (26.7%) while the rest of monoterpenes displayed a weak or no mortality after one week. (-)-Carvone, cuminaldehyde and geraniol caused the highest mortality at the rate of 1 mg/g with mortality percentages of 96.7, 88.3 and 85.0, respectively. At the rate of 5 mg/g, four of the tested monoterpenes (carvone, cuminaldehyde, geraniol and (-)-linalool) caused complete mortality to the adults of *S. oryzae* after one week. Despite their weak effect on the adult mortality at the rates of 0.5 and 1 mg/g after one week, (L)-fenchone and (-)-limonene showed a strong mortality of the rate of 5 mg/g with percentages of 91.7 and 88.3, respectively. In general,

(-)-carvone, cuminaldehyde, geraniol and (-)-linalool had a strong effect on the mortality of *S. oryzae* on treated wheat after one week of treatment. In contrast, myrcene and 1-8-cineole were the least effective monoterpenes on adult mortality. The results of *S. oryzae* adult mortality after two weeks indicated that there was a slight increasing in the mortality percentages comparing with those after one week.

In the case of T. castaneum, (-)-carvone was the only compound which caused a significant mortality (61.7%) at the rate of 0.5 mg/g., while the other monoterpenes showed a weak or no effect on mortality after one week of treatment (Table 2). (-)-carvone, cuminaldehyde and geraniol were the most toxic at the rate of 1 mg/g. All of the tested monoterpenes except myrcene and (-)-linalool caused complete mortality of T. castaneum adults after one week at the rate of 5 mg/g. The mortality of T. castaneum adult caused by myrcene, geraniol, (-)-linalool and cuminaldehyde was significantly increased after two weeks comparing with that of one week. The toxicity results of monoterpenes on the adults of S. oryzae and T. castaneum revealed that (-)-limonene, (L)-fenchone and cuminaldehyde had approximately the same activity on both insects. Myrcene, geraniol, (-)-carvone and 1-8-cineole were more toxic towards T. castaneum than S. oryzae. In contrary, (-)-linalool was more toxic towards S. oryzae than T. castaneum. The different response of these two insects to monoterpenes has been stated (Lee et. al., 2003 and 2004).

The investigation of a relationship between the toxicities of the test monoterpenes and their structures revealed some interesting correlations. For examples, (-)-carvone, a ketone, showed the highest toxicity against the two tested insects. Cuminaldehyde, an aldehyde, and geraniol, an alcohol, were more toxic than other oxygenated and non-oxygenated monoterpenes. On the other hand, 1-8-cineole, an ether, and (-)-limonene, a hydrocarbon, showed a moderate toxicity against both insects. Myrcene, a hydrocarbon, was the less effective monoterpene among the test compounds against both insects. These findings are in agreement with Lee *et al.*, (2003) who stated that the two ketones, (-)-carvone and (L)-fenchone, were more effective fumigants than the alcohols, (-)-linalool and geraniol, and with Rice and Coats (1994) who found that some ketones were more effective fumigants than alcohols.

Table 2. Effect of monoterpenes aganist Tribolium castaneum exposed for

1 and 2 weeks on treated wheat.

Monoterpene	Monoterpene	Application rate (mg/g)	Cumulative mortality (% ± SE)	
class			1 Week	2 Weeks
-	Control	0	0.0 ± 0.0^{h}	0.0 ± 0.0^{g}
Hydrocarbon	(-)-Limonene	0.5	0.0 ± 0.0^{h}	1.7 ± 2.11^{8}
1170100010011	() =	1	$6.7 \pm 2.11^{\text{fgh}}$	13.3 ± 2.11
		5	100.0 ± 0.0^{n}	100.0 ± 0.0
	Мутсепе	0.5	0.0 ± 0.0^{h}	16.7 ± 2.11
	-	1	3.3 ± 2.11^{fgh}	21.7 ± 2.11
		5	$61.6 \pm 3.34^{\circ}$	63.3 ± 2.11
Alcohol	Geraniol	0.5	10.0 ± 0.0^{f}	31.7 ± 2.11
		1	48.3 ± 2.11^{d}	63.3 ± 2.11
		5	100.0 ± 0.0^{a}	100.0 ± 0.0
	(-)-Linalool	0.5	$5.0 \pm 0.04^{\text{fgh}}$	21.7 ± 3.34
	() =	1	$16.7 \pm 3.34^{\circ}$	21.7 ± 2.11
		5	51.7 ± 3.34^{d}	63.3 ± 4.41
Ketone	(-)-Carvone	(.5	$61.7 \pm 3.34^{\circ}$	65.0 ± 2.89
reione	()	1	$100.0 \pm 0.04^{\text{a}}$	100.0 ± 0.00
		5	100.0 ± 0.0^{a}	100.0 ± 0.0
	(L)-Fenchone	0.5	1.7 ± 2.11^{gh}	3.3 ± 2.11
	(2) 1 0	1	8.3 ± 2.11^{fg}	11.7 ± 2.11
		5	100.0 ± 0.0^{a}	100.0 ± 0.0
Aldehyde	Cuminaldehyde	0.5	$16.7 \pm 2.11^{\circ}$	25.0 ± 2.89
	<i></i>	1	76.7 ± 3.34^{b}	96.7 ± 3.34
		5	100.0 ± 0.0^{a}	100.0 ± 0.0
Oxide	1-8-Cineole	0.5	0.0 ± 0.0^{h}	1.7 ± 2.11
		1	8.3 ± 2.11^{fg}	21.7± 2.11
		5	100.0 ± 0.0^a	$100.0 \pm 0.$

Means in a column with same letters are not-significant (p < 0.05) compared to the control.

Effect of monoterpenes on progeny production of S. oryzae: The results of the progeny production and the reduction percentage of S. oryzae after 6 and 12 weeks of treatment are presented in Table 3. In general, all of the tested compounds except for myrcene showed significant reduction in progeny compared with control. (-)-Carvone was most effective in reducing the progeny production at the three tested application rates, followed by geraniol, cuminaldehyde and (-)-linalool. No progeny emerged on wheat treated with these four monoterpenes at the highest application rate of 5 mg/g. Two monoterpenes ((L)-fenchone and (-)-limonene) revealed a strong reduction in progeny at 5 mg/g, while 1-8-cineole showed a moderate progeny reduction. In contrast, myrcene was the less effective compound;

there were no significant differences in progeny reduction at the three tested rates of this compound compared with control.

Table 3. Effect of monoterpenes on the progeny production (±SE) of

Sitophilus oryzae on treated wheat.

$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	Ditopinius Ory.	ede on ticato	T	-le-	1	
$ \begin{array}{ c c c c c c } \hline \textbf{Nonoterpene} & \textbf{rate (mg/g)} & \textbf{Progeny} \\ \hline \textbf{production} \\ \hline \textbf{Control} & \textbf{0} & 165.0\pm5.23^{ab} \\ \hline \textbf{(-)-Limonene} & 0.5 & 163.0\pm4.05^{ab} \\ \hline 1 & 131.0\pm1.53^{d} & 20.6\pm0.92 & 336.0\pm10.98^{g} \\ \hline \textbf{5} & 14.7\pm1.20^{gh} & 91.1\pm0.72 & 43.7\pm1.33^{m} & 92.2\pm0.23 \\ \hline \textbf{Myrcene} & 0.5 & 170.3\pm5.21^{b} & -0.83\pm3.85 & 580.04.93^{a} & -3.3\pm0.90 \\ \hline \textbf{1} & 164.0\pm7.01^{ab} & 0.6\pm4.26 & 562.0\pm4.05^{b} \\ \hline \textbf{5} & 161.3\pm7.23^{ab} & 2.4\pm3.68 & 559.0\pm3.61^{b} \\ \hline \textbf{Geraniol} & 0.5 & 127.0\pm4.05^{d} & 23.0\pm2.46 & 265.0\pm4.62^{i} & 52.8\pm0.81 \\ \hline \textbf{1} & 13.0\pm2.0^{gh} & 92.1\pm1.20 & 31.0\pm3.06^{m} & 94.4\pm0.49 \\ \hline \textbf{5} & 0.0\pm0.0^{h} & 100.0\pm0.0 & 0.0\pm0.0^{n} & 100.0\pm0.0 \\ \hline \textbf{(-)-Linalool} & 0.5 & 139.0\pm3.79^{cd} & 15.8\pm2.30 & 395.0\pm7.10^{f} & 29.6\pm1.27 \\ \hline \textbf{1} & 53.0\pm7.20^{f} & 67.9\pm4.40 & 195.0\pm5.04^{k} & 65.3\pm0.89 \\ \hline \textbf{5} & 0.0\pm0.0^{h} & 100.0\pm0.0 & 0.0\pm0.0^{n} & 100.0\pm0.0 \\ \hline \textbf{(-)-Carvone} & 0.5 & 85.7\pm7.27 & 48.1\pm4.53 & 246.7\pm5.85^{i} & 56.0\pm1.23 \\ \hline \textbf{1} & 3.3\pm0.34^{gh} & 98.0\pm0.20 & 30.0\pm0.58^{m} & 94.7\pm0.08 \\ \hline \textbf{5} & 0.0\pm0.0^{h} & 100.0\pm0.0 & 0.0\pm0.0^{n} & 100.0\pm0.0 \\ \hline \textbf{(L)-Fenchone} & 0.5 & 153.0\pm5.04^{bc} & 7.2\pm3.06 & 506.7\pm9.85^{cd} & 9.7\pm1.77 \\ \hline \textbf{1} & 132.0\pm3.0^{d} & 20.0\pm1.80 & 495.0\pm3.47^{de} \\ \hline \textbf{5} & 4.0\pm0.58^{gh} & 97.6\pm0.35 & 28.3\pm3.52^{m} & 95.0\pm062 \\ \hline \textbf{Cuminaldehyde} & 0.5 & 149.0\pm3.06^{bc} & 10.5\pm1.44 & 487.7\pm6.97^{e} & 13.1\pm1.25 \\ \hline \textbf{1} & 19.0\pm0.58^{g} & 88.5\pm0.35 & 79.7\pm5.931 & 85.8\pm1.07 \\ \hline \end{array}$	Manatarnana	Application	6 weeks		12 weeks	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	Monoterbene			Reduction		Reduction
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$						Reddenon
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$		_				
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	(-)-Limonene	0.5			521.3±6.39°	7.1±1.16
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$]]		20.6±0.92	336.0±10.98 ^g	40.1±1.95
Geraniol $\begin{array}{c ccccccccccccccccccccccccccccccccccc$				91.1±0.72	43.7±1.33 ^m	92.2±0.23
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	Myrcene	0.5		-0.83±3.85	580.04.93ª	-3.3±0.90
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	1	1		0.6±4.26	562.0±4.05 ^b	-0.1±0.69
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		ì		2.4±3.68	559.0±3.61b	0.43±0.66
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	Geraniol	0.5		23.0±2.46	265.0±4.62 ⁱ	52.8±0.81
(-)-Linatool 0.5 139.0 ± 3.79^{cd} 15.8 ± 2.30 395.0 ± 7.10^f 29.6 ± 1.27 153.0 ± 7.20^f 67.9 ± 4.40 195.0 ± 5.04^k 65.3 ± 0.89 0.0 ± 0.0^h 100.0 ± 0.0 0.0 ± 0.0^n 100.0 ± 0.0 $100.0\pm0.$		1		92.1±1.20	31.0±3.06 ^m	94.4±0.49
(-)-Carvone (1) $\begin{array}{cccccccccccccccccccccccccccccccccccc$				100.0±0.0	0.0±0.0 ⁿ	100.0±0.0
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	(-)-Linalool	0.5		15.8±2.30	395.0±7.10 ^f	29.6±1.27
(-)-Carvone 0.5 85.7 ± 7.27 48.1 ± 4.53 $246.7\pm5.85^{\rm j}$ 56.0 ± 1.23 $3.3\pm0.34^{\rm gh}$ 98.0 ± 0.20 $30.0\pm0.58^{\rm m}$ 94.7 ± 0.08 $0.0\pm0.0^{\rm h}$ 100.0 ± 0.0 0.0 ± 0.0 0.0 ± 0		1		67.9±4.40	195.0±5.04k	65.3±0.89
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$			0.0±0.0 ^h	100.0±0.0	0.0 ± 0.0^{n}	100.0±0.0
(L)-Fenchone $ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	(-)-Carvone	0.5		48.1±4.53	246.7±5.85 ^j	56.0±1.23
(L)-Fenchone $ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		1	3.3±0.34gh	98.0±0.20	30.0±0.58 ^m	94.7±0.08
(L)-Fenchone 0.5 153.0 ± 5.04^{bc} 7.2 ± 3.06 506.7 ± 9.85^{cd} 9.7 ± 1.77 132.0 ± 3.0^{d} 20.0 ± 1.80 495.0 ± 3.47^{de} 11.8 ± 0.64 4.0 ± 0.58^{gh} 4.0 ± 0.58^{gh} 4.0 ± 3.06^{bc} 4.0		5	0.0±0.0 ^h	100.0±0.0	0.0±0.0"	
Cuminaldehyde	(L)-Fenchone	0.5	153.0±5.04 ^{bc}	7.2±3.06	506.7±9.85 ^{cd}	
Cuminaldehyde 0.5 149.0 ± 3.06^{bc} 10.5 ± 1.44 487.7 ± 6.97^{c} 13.1 ± 1.25 19.0 ± 0.58^{g} 88.5 ± 0.35 79.7 ± 5.931 85.8 ± 1.07	•	1		20.0±1.80	495.0±3.47 ^{de}	11.8±0.64
1 19.0±0.58 ^g 88.5±0.35 79.7±5.931 85.8±1.07		5		97.6±0.35	28.3±3.52 ^m	95.0±062
, and and	Cuminaldehyde	0.5	149.0±3.06 [№]	10.5±1.44	487.7±6.97°	13.1±1.25
5 0.0+0.0h 100.0+0.0 0.0+0.0h 100.0+0.0		1	19.0±0.58 ^g	88.5±0.35	79.7±5.931	85.8±1.07
	,	5	0.0±0.0 ^h	100.0±0.0	0.0 ± 0.0^{n}	100.0±0.0
1-8-Cineole 0.5 139.0 ± 4.17^{cd} 15.7 ± 2.53 397.0 ± 8.63^{f} 29.3 ± 1.54	1-8-Cineole	0.5	139.0±4.17 ^{cd}	15.7±2.53	_	l
1 130.3 ± 4.49^{d} 21.0 ± 3.32 380.0 ± 6.09^{f} 32.3 ± 1.10		1	130.3±4.49 ^d	21.0±3.32		32.3±1.10
			95.7±1.86°	42.0±1.11		

Means in a column with same letters are not-significant (p < 0.05) compared to the control.

Progeny production in the treated substrate is perhaps more important than parental mortality, because a grain protectant should protect the grain for a long storage period (Athanassiou et al., 2005). In our work, progeny production of S. oryzae was significantly low on wheat treated with 1 and 5 mg/g of the tested monoterpenes except for myrcene, which suggest that long-term protection for the treated wheat.

Effect of monoterpenes on weight loss of wheat: All of the tested monoterpenes except myrcene reduced significantly weight loss of treated wheat particularly at the higher rates of 1 and 5 mg/g (Table 4). Complete

protection was observed at the application rate of 5 mg/g in wheat treated with (-)-carvone, geraniol, cuminaldehyde and (-)-linalool. At the application rate of 1 mg/g, (-)-carvone, cuminaldehyde and geraniol showed a strong reduction of weight loss with percentages of loss of 0.8, 1.9 and 4.9%, respectively. The reduced percentage of grain damage observed in the treated grains with the application rates of 1 and 5 mg/g can be attributed to high mortality of adults after treatment with the tested monoterpenes.

Table 4. Effect of monoterpenes on weight loss (%± SE) of treated wheat.

Monoterpene	Application rate (mg/g)	Weight loss (gm)	Monoterpene	Application rate (mg/g)	Weight loss (gm)
Control	0	34.1±1.22ab	(-)-Carvone	0.5	16.3 ±1.24 ^f
(-)-Limonene	0.5	36.0 ±1.80°	•	1	0.8 ± 0.0^{8}
	1	28.5 ± 1.10^{cde}		5	0.0 ± 0.0^{g}
	5	2.4 ± 0.69^8	(L)-Fenchone	0.5	35.6 ±1.47°
Myrcene	0.5	$33.5 \pm 2.06^{\text{abc}}$		1	31.3 ± 0.78^{abcd}
]	33.3 ± 1.15^{abc}		5	1.3 ± 0.30^{g}
	5	32.0 ± 0.95^{abc}	Cuminaldehyde	0.5	29.9 ± 0.34^{bcde}
Geraniol	0.5	$24.6 \pm 0.35^{\circ}$	·	1	4.7±1.418
	1	1.9 ± 0.71^8		5	0.0 ± 0.0^{g}
	5	0.0 ± 0.0^{g}	1-8-Cincole	0.5	26.6 ±1.68°
(-)-Linalool	0.5	25.1±1.54°		1	27.1±1.21dc
	1	16.9 ± 2.31^{f}		5	24.9 ±1.65°
	5	0.0 ± 0.0^8			

Means in a column with same letters are not-significant (p < 0.05) compared to the control.

Effect of monoterpenes on seed germination of treated wheat: Seed germination results revealed that the tested monoterpenes had no and/or slight effect on germination percentage at the highest rate of 5 mg/g after 12 weeks of treatment (Figure 2). The untreated seeds (control) were completely destroyed therefore no seed germination was observed. Cuminaldehyde and (-)-linalool had no effect on seed germination with germination percentage of 100 % for each one, while (-)-carvone and (L)-fenchone had slight effect on seed germination with germination percentages of 95.0 and 93.3%, respectively. In contrary, (-)-limonene and (-)-limonene showed significant effect on seed germination with seed germination percentages of 76.7 and 83.3, respectively.

The fun-igant and contact toxicities of monoterpenes against S. oryzae and T. castaneum have been investigated (Lee et al. 2003, 2004; Park et al., 2003; Abdelgaleil et al. 2009). However, there were no reported studies, to the best of our knowledge, on the effectiveness of monoterpenes for control of these two insects on the stored wheat or other stored products. The results of the present work revealed that some of the tested monoterpenes had a

potential as seed protectants. Since, they caused high mortality, reduced progeny and protected wheat seeds free of infestation for 12 weeks.

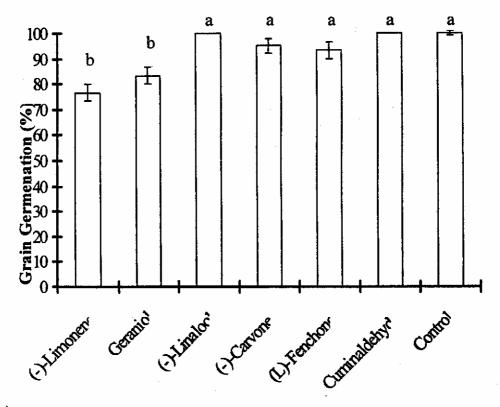


Figure 2. Effect of monoterpenes on grain germination of wheat treated with the application rate of 5 mg/g,12 weeks after treatment.

Although synthetic insecticides and/or furnigants are the most widely used methods for managing stored-product insects, there are some limitations for their use in control such as the development of resistance, residues in stored products and environmental pollution (Brun and Attia, 1983; Collins et al., 1993; Lorini and Galley, 1999; Zettler and Arthur, 2000). Replacement of methyl bromide, the most effective furnigant for insect disinfestation in storage facilities, is essential because it depletes the ozone layer (UNEP, 1995) and thus searching for new compounds with new mode of action and environmentally safe are required. It should be noted that biologically active compounds of food plants such as monoterpenes are assumed to be

environmentally more acceptable and less hazardous than others to humans. The results of the present study demonstrated that some of tested monoterpenes such as (-)-carvone, geraniol, cuminaldehyde and (-)-linalool had remarkable toxicity against *S. oryzae* and *T. castaneum*. Moreover, these monoterpenes drastically reduced the progeny *S. oryzae* and the damage of wheat seeds. Based on these results, these monoterpenes could be used as safer pest control agents against *S. oryzae* and *T. castaneum*.

REFERENCES

- Abdelgaleil, S.A.M.; Mohamed, M.I.E.; Badawy, M.E.I. and El-arami, S.A.A. (2009). Fumigant and contact toxicities of monoterpenes to *Sitophilus oryzae* (L.) and *Tribolium castaneum* (Herbst) and their inhibitory effects on acetylcholinesterase activity. J. Chem. Ecol., 35: 518-525.
- Argandoña, V.H.; Rovirosa, J.; San-Martín, A.; Riquelme, A.; Díaz-Marrero, A. R.; Cueto, M.; Darias, J.; Santana, O.; Guadaño, A. and González-Coloma, A. (2002). Antifeedant effects of marine halogenated monoterpenes. J. Agric. Food Chem., 50: 7029 -7033.
- Athanassiou, C.G.; Kavallieratos, N.G.; Economou, L.P.; Dimizas, C.B. and Vayias, B.J. (2005). Persistence and efficacy of three diatomaceous earth formulations against *Sitophilus oryzae* (Coleoptera: Curculionidae) on wheat and barley. J. Econ. Entomol., 98:1404–1412.
- Brun, L.O. and Attia, F.I. (1983). Resistance to lindane, malathion and fenitrothion in coleopterous pests of stored products in New Caledonia. Proceedings of the Hawaiian Entomological Society, 24: 211-215.
- Choi, W.-S.; Park, B.-S.; Lee, Y.-H.; Jang, D.Y.; Yoon, H. Y. and Lee, S.-E. (2006). Furnigant toxicities of essential oils and monoterpenes against *Lycoriella mali* adults. Crop Protect., 25: 398-401.
- Collins, P.J.; Lambkin, T.M.; Bridgeman, B.W. and Pulvirenti, C. (1993). Resistance to grain protectant insecticides in oleopterous pests of stored cereals in Queensland, Australia. J. Econ. Entomol., 86: 239-245.

- J. Pest Cont. and Environ. Sci. 17 (1/2): 61-76 (2009)
- Evans, D.E. (1987). Stored products. In: Burn, A.J., Coaker, T.H., Jepson, P.C. (Eds.), Integrated Pest Management. Academic Press, London, UK, pp. 425-461.
- Gunderson, C.A.; Samuelian, J.H.; Evans, C.K. and Brattsten, L.B. (1985). Effects of the mint monoterpene pulegone on *Spodoptera eridania* (Lepidoptera: Noctuidae). Environ. Entomol., 14: 859–863.
- Hough-Goldstein, J.A. (1990). Antifeedant effects of common herbs on the Colorado potato beetle (Coleoptera: Chrysomelidae). Environ. Entomol., 19: 234–238.
- Hummelbrunner, L.A. and Isman M.B. (2001). Acute, sublethal, antifeedant, and synergistic effects of monoterpenoid essential oil compounds on the tobacco cutworm, *Spodoptera litura* (Lep., Noctuidae). J. Agric. Food Chem., 49: 715-720.
- Isman, M.B. (2000). Plant essential oils for pest and disease management. Crop Protect., 19: 603-608.
- Karr, L.L. and Coats, J.R. (1992). Effects of four monoterpenoids on growth and reproduction of the German cockroach (Blattodea: Blattellidae). J. Ecol. Entomol., 85: 424-429.
- Kim, D.H. and Ahn, Y.J. (2001). Contact and fumigant activities of constituents of *Foeniculum vulgare* fruit against three Coleopteran stored-product insects. Pest Manag. Sci., 57: 301-306.
- Kohli, R.K.; Batish, D. and Singh, H. P. (1998). Eucalypt oils for the control of parthenium (*Parthenium hysterophorus* L.). Crop Protect., 17: 119–122.
- Kordali, S.; Aslan, I.; Calmasur, O. and Cakir, A. (2006). Toxicity of essential oils isolated from three *Artemisia* species and some of their major components to granary weevil, *Sitophilus granarius* (L.) (Coleoptera: Curculinonidae). Ind. Crops Prod., 23: 162-170.
- Larry, P.P. (2000). Entomology and Pest Management, Fourth Ed. Prentice-Hall, India.

- Lee, B.-H.; Annis, P. C.; Tumaaliia, F.and Choic W.-S. (2004). Fumigant toxicity of essential oils from the Myrtaceae family and 1,8-cineole against 3 major stored-grain insects. J. Stored Prod. Res., 40: 553-564.
- Lee, B.-H.; Choi, W.-S.; Lee S.-E.and Park, B.-S. (2001). Fumigant toxicity of essential oils and their constituent compounds towards the rice weevil, Sitophilus oryzae (L.) Crop Protect., 20: 317-320.
- Lee, S.; Peterson C. J. and Coats, J.R. (2003). Fumigation toxicity of monoterpenoids to several stored product insects. J. Stored Prod. Res., 39: 77-85.
- Lorini, I. and Galley, D.J. (1999). Deltamethrin resistance in *Rhyzopertha dominica* (F.) (Coleoptera: Bostrichidae), a pest of stored grain in Brazil. J. Stored Prod. Res., 35: 37-45.
- Mason, J.R. (1990). Evaluation of *d*-pulegone as an avian repellent. J. Wildlife Manag., 54: 130-135.
- Mohan, S. and Fields, P.G. (2002). A simple technique to assess compounds that are repellents or attractive to stored product insects. J. Stored Prod. Res., 33: 289-298.
- Park, I.K.; Lee, S.G.; Choi, D.H.; Park, J.D. and Ahn, Y.J. (2003). Insecticidal activities of constituents identified in the essential oil from leaves of *Chamaecyparis obtusa* against *Callosobruchus chinensis* (L.) and *Sitophilus oryzae* (L.). J. Stored Prod. Res., 39: 375-384.
- Prates, H.T.; Santos, J.P.; Maquil, T.M.; Fabris, J.D.; Oliveira A.B. and Foster J.E. (1998). Insecticidal activity of monoterpenes against *Rhyzopertha dominica* (F.) and *Tribolium castaneum* (Herbst). J. Stored Prod. Res., 34: 243-249.
- Rice, P.J. and Coats, J.R. (1994). Insecticidal properties of several monoterpenoids to the house fly (Diptera: Muscidae), red flour beetle (Coleoptera: Tenebrionidae), and southern maize rootworm (Coleoptera: Chrysomelidae). J. Econ. Entomol., 87: 1172-1179.

- J. Pest Cont. and Environ. Sci. 17 (1/2): 61-76 (2009)
- Romagni, J.G.; Allen, S.N. and Dayan, F.E., 2000. Allelopathic effects of volatile cineoles on two weedy plant species. J. Chem. Ecol., 26: 303–313.
- Samarasekera R.; Weerasinghe I.S. and Hemalal K.P. (2008). Insecticidal activity of menthol derivatives against mosquitoes. Pest Manag. Sci., 64: 290-295.
- Templeton, W. (1969). An introduction of chemistry of terpenoids and steroids. Butterworths, London.
- UNEP (1995). Montreal protocol on substances that deplete the ozone layer.1994 Report of the Methyl Bromide Technical Options Committee.1995 Assessment. United Nations Environmental Programme, Nairobi, Kenya, 304pp.
- Waliwitiya R.; Isman M.B.; Vernon R.S. and Riseman, A. (2005). Insecticidal activity of selected monoterpenoids and rosemary oil to *Agriotes obscurus* (Coleoptera: Elateridae). J. Econ. Entomol., 98: 1560-1565.
- Watanabe, K.; Shono, Y.; Kakimizu, A.; Okada, A.; Matsuo, N.; Satoh, A. and Nishimura, H. (1993). New mosquito repellent from *Eucalyptus camaldulensis*. J. Agric. Food Chem., 41: 2164-2166.
- Windholz, M.; Budavari, S.; Blumetti, R. F. and Otterbein, E. S. (1983). The Merck index. Merck, Rahway, NJ.
- Wink, M. (1993). Production and application of phytochemicals from an agricultural perspective. In: Van Beek, T.A., Breteler, H. (Eds.), Phytochemistry and Agriculture, Vol. 34. Clarendon, Oxford, UK, pp. 171-213.
- Zettler, J.L. and Arthur, F.H. (2000). Chemical control of stored product insects with fumigants and residual treatments. Crop Protect., 19: 577-582.

فاعلية المونوتربينات في مكافحة حشرتي سوسة الأرز وحنفساء الدقيق الصدنية في القمح المخزون

سيلان أحمد العرامي - مجدى إبراهيم الدسوقي - سمير عبد العظيم عبد الجليل

تم إجراء هذه الدراسة لتقييم فاعلية ثمانية من المونوتربينات وهي carvone-(-) و-8-1-(-)-limonene (-)- geraniol (L)-fenchone cuminaldehyde scineole linalool ضد حشرتين رئيسيتين من حشرات الحبوب المخزونة على حبوب القمح معملياً. الحشرات التي تمت الدراسـة عليهـا هـي سوسـة الارز وخنفساء الدقيق الـصدنية. تم معاملـة حبوب القمح بالمعدلات 0.5 و 1 و 5 مجم/جم لمكافحة الحشرتين تحت الإختبار. الـ carvone-(-) كان أكثر المركبات سمية ضد الحشرتين على كل المعدلات المستخدمة بعد اسبوع واسبوعين من المعاملة. الـ geraniol و cuminaldehyde كانا ضمن أكثر المركبات فاعلية ضد الحشرتين. أربعة من المونوتربينات المختبرة وهي الـ cuminaldehyde و geraniol و cuminaldehyde linalool -(-) قد سببت موت كامل لحشرة سوسة الأرز بعد اسبوع من المعاملة على معدل 5 مجم/جم في حين كل المونوتربينات المختبرة عدا myrcene و linalool -(-) سببت موت كامل لمشرة خنفساء الدقيق الصدنية على نفس معدل الإستخدام. بمقارنية سمية المونوتربينات ضد الحشرتين تحت الدراسة وجد أن الـ L)- fenchone (-) و cuminaldehyde) لهما نفس الفاعلية ضد المشرتين تقريباً. على الجانب الأخر geraniol و myrcene و -(-) carvone و 1-8-cineole كانت أكثر فاعلية على خنفساء الدقيق الصدنية من سوسة الأرز بينما الـ linalool-(-) كان أكثر سمية ضد سوسة الأرز من خنفساء الدقيق الصدنية. كل المونوتربينات المختبرة عدا الـ myrcene سببت خفض معنوى في أعداد حشرات سوسة الأرز الناتجة بعد 6 و12 اسبوع من المعاملة مقارنة بالكنترول. الـ carvone-(-) كان أعلى المركبات فاعلية في خفض أعداد الحشرات الناتجة على الثلاث معدلات المستخدمة يليه الـ geraniol و cuminaldehyde و -(-) linalool . المركبات الأربعة السابقة منعت تماماً وجود حشرات على معدل 5 مجم/جم بعد 12 اسبوع. على معدلات 1 و5 مجم/جم غالبية المونوتربينات قللت الفقد في الوزن لحبوب القمح المعاملة وكذلك أظهرت مركبات الـ carvone-(-) و geraniol و cuminaldehyde و -(-) linalool وقايـة كاملـة للحبوب على معدل 5 مجم/جم. هذا النشاط الإبـادي القوى للمونوتربينـات خاصة الـ cuminaldehyde و geraniol و cuminaldehyde و linalool -(-) يدل على أن هذه المونوتربينات يمكن أن تستخدم في مكافحة حشرتي سوسة الأرز و خنفساء المدقيق الصدئية في