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Fumigation of Volatile Monoterpenes and Aromatic Compounds Against Adults of *Sitophilus granarius* (L.) (Coleoptera: Curculionidae)

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Abstract: In the present study, 42 pure monoterpenes and volatile aromatic compounds were tested to evaluate their toxicities against adults of granary weevil, *Sitophilus granarius* (L.) (Coleoptera: Curculionidae) at laboratory condition. For the insecticidal activity tests, 10 and 20 µL/Petri dish concentrations for liquid compounds and 10 and 20 µg/Petri dish concentrations for solid compounds were used. The results show that majority of tested compounds were found to be toxic against to the adults of *S. granarius* and they showed various degrees of toxicity with range of 6–100% mortality. Among the compounds tested, oxygenated monoterpenes, linalool, carvacrol, terpinen-4-ol, limonene oxide, carvone, dihydrocarvone, fenchone, menthone and aromatic volatiles, *p*-anis aldehyde, benzyl acetate, cinnamyl aldehyde in the current study displayed complete mortality on the pest adults after 24h of exposure. In general, oxygenated monoterpenes and aromatic compounds exhibited high toxicity as compared with monoterpene hydrocarbons. Among the monoterpenes hydrocarbons, the highest toxic effects were shown by *γ*-terpinene, limonene and *β*-pinene. Our results showed that linalool, carvacrol, terpinen-4-ol, limonene oxide, carvone, dihydrocarvone, fenchone, menthone, *p*-anisaldehyde, benzyl acetate and cinnamyl aldehyde were the most toxic compounds against the adults of the pest. Therefore, these compounds can be used as natural insecticides against the *S. garanarius* adults.

Keywords: Toxicity; *Sitophilus granarius;* volatiles; monoterpenes; carvacrol; linalool; terpinen-4-ol.

1. Introduction

Sitophilus granarius (L.) (Coleoptera: Curculionidae) known as "the wheat weevil", "the grain weevil or "granary weevil is an important pest widely distributed in the world. It is a common pest in many places of the world. It cause important damage to stored grains, wheat, rye, barley, rice and corn and can sorely decrease crop yields of stored grain products. Hence, *S. granarius* causes large economic losses of stored wheat grains. It is difficult to detect the amount of its damages worldwide and in general, all of the grain in an infected storage facility should be destroyed [1-3]. Every day, the grain needs are increasing in the world depending on the world's artistic population. For this reason, it is of great importance the control of the wheat weevil to reduce its harmful effects.

Several insecticides have been tried for years to prevent this damage. Synthetic chemical insecticides and fumigants are frequently used for the control of this pest [1-3]. Nowadays, the conventional ways to control of this insect are applying the insecticides, either directly applied to

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grains or by gas fumigation. However, new strategies for pest and disease control to be used in rotation with or replacement of conventional pesticides are required in the pest management. Synthetic pesticides also lead to the environmental pollution because of their slow biodegradation [1-3]. Additionally, the risks of the microorganism's resistance and the high ratio of cost–benefit of synthetic pesticide consumption are other disadvantages [4,5]. These disadvantage sides of synthetic pesticides have created a viewpoint for introduction of alternative pesticides [6]. Harmful effects of natural substances to mammalians health and the environment are relatively less than those of synthetic chemicals. Therefore, there is a considerable interest in developing natural pesticides, additional fumigants and control measures as alternatives to non-selective synthetic pesticides to control the pests of medical and economic importance [7-11]. Over the past 15 years, interest in botanical insecticides has increased as a consequence of environmental concerns and insect populations becoming resistant to conventional chemicals. Natural insecticides are inherently synthesized metabolites as defense agents of the plant kingdom [12]. Nowadays, natural insecticides constitute 1% of the world insecticide market [13]. In spite of the common recognition that many plant species possess insecticidal properties, solely a handful of herbal pest control agents are in use due to commercialization problems of new insecticides [14,15]. Some secondary metabolites synthesized by plants play an important role in plant-insect interaction, and are commonly responsible for plant resistance to insects [16]. There were many reports concerning with the development of new alternative pesticides, such as insect growth regulators, fungal pathogens, toxic natural products including plant essential oils, extracts and secondary metabolites for pest control in agriculture [12, 17-27]. Accordingly, contact and fumigant insecticidal activities of natural pest and pesticides including plant essential oils and monoterpenes, which are important constituents of plant essential oils against stored product pests have been addressed [28-37]. It has also been demonstrated that pure monoterpenes are non-persistent in soil and water [38] and essential oils and their pure metabolites provide potential resources to develop new alternative pesticide, which less toxic to living health and environment.

Essential oils are native plant products that contain native flavors and fragrances grouped as monoterpenes (hydrocarbons and oxygenated derivatives), sesquiterpenes (hydrocarbons and oxygenated derivatives), aliphatic compounds (alkanes, alkenes, ketones, aldehydes, acids and alcohols) and aromatic compounds that provide characteristic odors. Insecticidal properties of numerous essential oils and their monoterpenes components have been widely studied against to various insect species [29, 34, 36, 37, 39-48]. Monoterpenes possess acute contact and fumigant toxicity to various insects [41, 47, 49-53], repellent activity [54, 55], antifeedant activity [56, 57], as well as development and growth inhibitory activity [58, 59]. The aim of present work was to determine the fumigant effects of 42 pure monoterpenes and aromatic compounds against *S. granarius* adults.

2. Materials and methods

2.1. Pure monoterpenoids and chemicals

Pure volatile compounds were purchased commercially from Fluka, Sigma, Merck, Aldrich and Alfa Company. Their chemical structures are shown in Fig. 1. The compounds tested for toxicity against *S. granarius* were anethole (Fluka, purity 98%), *p*-anisaldehyde (Sigma, purity 97%), benzyl acetate (Fluka, purity 99%), borneol (Fluka, purity 95%), bornyl acetate (Sigma, purity 97%), camphene (Fluka, purity 90–95%), carvacrol, camphor (Fluka, purity 97%), carvone (Fluka, purity 99%), 3-carene (Aldrich, purity 95%), *β*-citronellene (Fluka, purity 95%), *β*-citronellol (Fluka, purity 95%), cinnamyl alcohol (Fluka, purity 97%), cinnamyl aldehyde (Fluka, purity 98%), 1,8-cineole (Sigma, purity 98%), cumin aldehyde (Fluka, purity 90-95%), dihydrocarvone (Alfa, purity 98%), eugenol (Fluka, purity 99%), ethyl cinnamate (Fluka, purity 98%), fenchol (Fluka, purity 98%), fenchone (Fluka, purity 98%), geraniol acetate (Alfa, purity 97%), quaiazulene (Fluka, purity 98%), isoeugenol (Fluka, purity 95%), isomenthol (Alfa, purity 99%), linalool acetate (Fluka, purity 95%), linalool (Fluka, purity 97%), limonene (Fluka, purity 98%), limonene oxide (Aldrich, purity 97%), menthol (Fluka, purity 99%), menthone (Fluka, purity 98%), methyl cinnamate (Fluka, purity 99%), myrcene (Aldrich, purity 98%), nerol (Sigma, purity 98%), neryl acetate (Alfa, purity 98%), *α*-pinene (Fluka, purity 95–97%), *β*-pinene (Fluka, purity 95%), thymol (Fluka, purity 99%), *γ*-terpinene (Sigma, purity 95%), *α*-terpineol (Merck, purity 98%), *p*-vanillin (Fluka, purity 98%) and DDVP (2, dichlorovinyl dimethyl phosphate) (Bayer, Turkey).

2.2. Biological material

The insects were collected from a storage house. Wheat grains were purchased from a local market and stored in a freezer −20 °C to maintain freshness. Wheat for *S. granarius* washed in tap water, dried and heated at to prevent pre-infestation before using for the experiments. *S. granarius* adults were reared in laboratory at 25 ± 1 °C, 64 ± 5 relative humidity and L:D=12 h:12 h in the Department of Plant Protection, Atatürk University, Erzurum-Turkey. The adults obtained from laboratory cultures were stored in separate insect cages containing enough of wheat. Tests are also carried out in under the same condition at the same laboratory.

2.3. Bioassays

To determine the toxicities of the compounds, 4-day-old adults of *S. granarius* were used as the insect material. In order to make test in the adult of the same age, some more larvae and pupa with wheat grain were placed in separate petri dishes. After adult emergence pupa, in the same time emergence adult were collected and used the same tests.

Glass Petri dishes (9 cm wide × 1.5 cm deep, corresponding to 120 mL volume) were used as exposure chambers to test the toxicities of pure commercial monoterpenes and aromatic compounds against adults of *S. granarius*. Ten microliters and 20 µL of liquid compounds were impregnated to Whatman no. 1 paper, which was stuck onto the top of Petri dishes from inside, by using an automatic pipette. The solid monoterpenes were solved in ethanol (1:1, w/v), and 20 and 40 µL of these solutions, corresponding to 10 mg and 20 mg/Petri dishes, respectively were impregnated to Whatman no. 1 paper in each Petri dish by using an automatic pipette. Ethanol was vaporized in atmospheric condition for 5 min. A filter paper was placed on bottom of each of Petri dishes (9 cm x 1.5 cm deep) and 20 adults of *S. granarius* were placed on this filter paper, containing the appropriate amounts (about 0.20 g) of wheat. Thus, there was no direct contact between the compounds and the adults. The Petri dishes were covered with a lid and transferred into incubator, and then kept under standard conditions of 25 ± 1 °C, 64 ± 5 relative humidity and 16:8 (light : dark) photoperiod for 4 days. Dichlorvos or 2,2-dichlorovinyl dimethyl phosphate (DDVP) (50 g/L), a commercial insecticide was used as positive control in the same conditions above mentioned. Ten microliters and 20 µL of positive control were applied, corresponding to 0.5 mg and 1 mg/Petri dishes, respectively. Control (sterile water and ethanol) treatments without the monoterpenes and aromatic compounds were treated in the same way. After exposure, the mortality of the adults was counted at 24^{th}, 48^{th}, 72^{th} and 96^{th} h. Grains were divided into two parts and in this way insects inside the grains became visible. At the end of each trial period, it was touched to the insects with forceps and observed movements of them. It was decided that completely inactive insects were dead. Toxicities of the monoterpenes and the aromatic compounds were expressed as % mean mortality of the adults in relation to the time 0. All experiments were carried out in triplicate at each dose.

2.4. Statistical analysis

The results of mean mortality were subjected to one-way variance analyses (ANOVA), using SPSS 17.0 software package. Mortality was expressed as mean (percentage) ± standard error. Differences between means were tested through LSD test and values with $p < 0.01$ were considered significantly different. In addition, durations for 50% and 90% mortality of the adults were determined in order to determine the toxic effects of substances used as insecticide by probit analysis.

3. Results and Discussion

In the present study, insecticidal effects of 31 pure commercial monoterpenes including monoterpene hydrocarbons and oxygenated monoterpenes and 11 aromatic compounds were determined against adults of *Sitophilus granarius.* The present results showed that the tested compounds have various toxic effects on the adults of *S. granarius* in comparison with control groups (ethanol, sterile water and DDVP) (Tables 1-3 and Figures S2-S5). Analysis of variance demonstrated that the effects of the monoterpenes and aromatic compounds on the mortality rate of S. *granarius* were more significant on the basis of both dosage rate and exposure time ($p < 0.05$). The toxicities of pure components on *S. granarius* increased with increase of dose and exposure times (Tables 1-3 and Figures S2-S5). Some oxygenated monoterpenes such as carvacrol, dihydrocarvone, fenchone, linalool, menthone, terpinen-4-ol, carvone, limonene oxide and aromatic compounds (*p*-anisaldehyde, benzyl acetate and cinnamyl aldehyde) exhibited potent toxic effects against the insect. After 24 h exposure, low dose (10 µL or 10 µg/Petri dish) of these compounds and positive control (DDVP) (0.1 mg/Petri dish) showed that complete mortality (Figures S2-S5). On the other hand, 1,8-cineole, *α*-terpineol, cumin aldehyde and methyl cinnamate were the other effective compounds for *S. granarius* adults among the tested compounds. The monoterpene hydrocarbons were found to be less toxic agents as compared with oxygenated monoterpenes and aromatic compounds (Table 1). Among the monoterpene hydrocarbons, *γ*-terpinene, limonene and *β*-pinene were the most toxic ones. As can be seen from Table 2, alcohols derivatives of oxygenated monoterpenes were also more toxic than their ester derivatives. For instance, two doses of linalool displayed %100 mortality after 24 h exposure, whereas its ester derivative, linalool acetate caused 44.4 % mortality after 24 h. exposure. Among the monoterpene hydrocarbons, camphene and α-pinene were the least effective agents against *S. granarius* adults in the dosage of 10 µL/Petri dish after 24 h (Tables 1 and 2, Figures S1 and S2). Furthermore, *p*-vanillin, guaiazulene and ethyl cinnamate were less effective compounds among the aromatic compounds (Table 3).

The durations, calculated by the probit analyses for the 50 and 90% mortality of the adults of the wheat weevil, *S. granarius* exposed to 10 µL or 10 µg of the pure monoterpenes and aromatic compounds were compiled in Table 4 [60]. The durations for the 50 and 90% mortality of linalool, carvacrol, terpinen-4-ol, limonene oxide carvone, dihydrocarvone, fenchone, menthone, *p*-anisaldehyde, benzyl acetate and cinnamyl aldehyde were not calculated due to 100% mortality of the wheat weevil after 24 h exposure of the treatments. These data showed that these compounds are potent toxic compounds against the adults of wheat weevil. Among other applications of monoterpenes and aromatic compounds, the shortest exposure times for 50 and 90% mortality of the pest were of cumin aldehyde, 1,8-cineole and bornyl acetate. The results presented in the Table 4 suggested that monoterpene hydrocarbons were the weak toxic reagents for the adults of the wheat weevil than oxygenated monoterpenes and aromatic compounds. On the other hand, the longest durations were observed for fenchol and isomenthol among the tested oxygenated monoterpenes (Table 2). Based on the durations for 50 and 90% mortality of the adults of wheat weevil, camphene was the least effective compound for the long duration times among the tested compounds.

Increasing use of natural insecticides will help to decrease the negative effects like toxicity to non-target animals, residue problems (environmental pollution) and insecticide resistance of chemical insecticides [61]. In this source, bio-insecticides may be also effective, bio-degradable and selective and associated with little advancement of resistance in the insect population and in the wake of, more safe to the environment. Previous reports demonstrated that in general, the toxicities of essential oils isolated from various plant samples against various stored pests were mainly related to their major components. These compounds in plant essential oils are generally described as monoterpenes [35, 47, 62, 63] and aromatic compounds [64]. In the current study, 42 pure volatiles including 8 monoterpenes hydrocarbons, 23 oxygenated monoterpenes and 11 aromatic compounds were tested for toxicities against the adults of wheat weevil, *S. granarius*. The current results indicated that the most effective compounds among the tested compounds were linalool, carvacrol, terpinen-4-ol, limonene oxide, carvone, dihydrocarvone, fenchone, menthone, *p*-anisaldehyde, benzyl acetate and cinnamyl aldehyde, with 100 % of mortality at low doses (24 h after treatment).

Table 1. Toxic effects of monoterpene hydrocarbons to adults of *S. granarius* (L.).

Treatments	Dose	Mean mortality (%)[a]			
		24[b]	48[b]	72[b]	96[b]
Controls					
Ethanol	10 µg	0.7± 2.6	2.0 ± 3.7	9.3 ± 4.7	10.6 ± 5.9
Sterile water	10 µg	0.0 ± 0.0	1.0 ± 0.8	3.1 ± 0.4	4.0 ± 0.8
DDVP	0.5 mg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	1 mg	100.0 ± 0.0*	100.0 ± 0.0	100.0 ± 0.0*	100.0 ± 0.0*
Monoterpenes hydrocarbons					
Camphene	10 µg	15.2 ± 1.7	20.2 ± 2.7*	22.2 ± 3.6*	27.3 ± 3.5*
	20 µg	20.2 ± 3.6*	38.4 ± 6.6*	45.5 ± 6.3*	49.5 ± 5.6*
3-Carene	8.7 µg	46.5 ± 2.7*	58.9 ± 2.7*	88.9 ± 2.0*	100.0 ± 0.0*
	17.4 µg	57.6 ± 3.0*	70.7 ± 5.3*	91.9 ± 1.0*	100.0 ± 0.0*
β-Citronellene	7.6 µg	18.2 ± 3.0*	42.4 ± 3.0*	62.6 ± 8.3*	87.9 ± 8.0*
	15.2 µg	25.3 ± 1.0*	58.6 ± 1.0*	77.8 ± 1.0*	93.9 ± 1.7*
Limonene	8.4 µg	33.3 ± 3.5*	70.7 ± 2.7*	91.9 ± 2.7*	96.0 ± 2.0*
	16.8 µg	92.9 ± 4.4*	97.0 ± 3.0*	100.0 ± 0.0*	100.0 ± 0.0*
Myrcene	7.9 µg	21.2 ± 1.7*	58.6 ± 3.6*	79.8 ± 2.0*	96.0 ± 2.0*
	15.8 µg	23.2 ± 2.7*	63.6 ± 4.6*	81.8 ± 3.5*	97.0 ± 2.0*
α-Pinene	8.6 µg	16.2 ± 2.0*	40.4 ± 2.0*	51.5 ± 1.7*	58.6 ± 1.0*
	17.2 µg	20.2 ± 1.0*	55.6 ± 14.6*	75.8 ± 8.0*	92.9 ± 4.4*
β-Pinene	8.7 µg	48.5 ± 5.2*	73.7 ± 5.1*	92.9 ± 4.4*	100.0 ± 0.0*
	17.4 µg	87.9 ± 1.7*	97.0 ± 3.0*	100.0 ± 0.0*	100.0 ± 0.0*
γ -Terpinene	8.5 µg	39.4 ± 1.7*	48.5 ± 3.5*	80.8 ± 2.7*	87.9 ± 3.5*
	17.0 µg	92.9 ± 3.6*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*

[a] Mean ± SE of three replicates, each set-up with 20 adults in relation to the time 0.
[b] Exposure time (h).
* Statistically different from negative controls (ethanol and sterile water) at p<0.05 according to LSD test.,

Other compounds that had strong fumigant insecticidal activity were 1, 8-cineole, *α*-terpineol, isomenthol, cumin aldehyde, bornyl acetate and methyl cinnamate according to their mortalities (%). These results suggested that the monoterpenes and aromatic compounds in essential oils might have different toxicity ratios and therefore, there are significant differences for the insecticidal effects of plant essential oils due to their different chemical constituents. In recent years, several studies were reported on the fumigation toxicity of some pure monoterpenoid constituents against the various insect species [34, 39, 40, 42, 44, 45, 47, 48, 65]. Kordali et al. (2006) tested the monoterpenes, borneol, bornyl acetate, camphor, 1,8-cineole, terpinen-4-ol and *α*-terpineol against *S. granarius* adults and in accordance with the present results, terpinen-4-ol, 1,8-cineole and *α*-terpineol were the most toxic compounds [46]. Likewise, 1,8-cineole, fenchone, linalool, terpinen-4-ol and menthone were found to be effective against the Colorado potato beetle [65]. Twenty monoterpenoids were evaluated in a preliminary fumigation toxicities test on important stored-product pests, *S. oryzae*, *Tribolium castaneum*, *Oryzaephilus surinamensis* and *Musca domestica* and *Blattella germanica* and 1,8-cineole, fenchone and pulegone was found to be most toxic agents against the pests [34]. Likewise, our results indicated that 1,8-cineole and fenchone were the effective compounds against the *S. granarius* (Table 2 and Figure S4).

A phenolic monoterpene, carvacrol is major and characteristic component of many essential oils of the species belonging to *Satureja*, *Origanum*, *Thymus* and *Thymbra* genus [27, 48, 66-70].

As can be seen from Table 2 and Figure S3, carvacrol was found to be more toxic compounds against the wheat weevil. In accordance with this result, it has been shown that carvacrol and essential oils contain mainly this compound have potent toxic effects against *S. granarius* and other insects [33, 48, 69, 71-74]. Furthermore, potent insecticidal effects of the monoterpenes, terpinen-4-ol [47, 69, 75] and carvone [77] against the adults of wheat weevil have been previously documented.

Table 2. Toxic effects of oxygenated monoterpenes to adults of *S. granarius* (L.).

Treatments	Dose	Mean mortality (%)[a]			
		24[b]	48[b]	72[b]	96[b]
Alcohols					
Borneol	10 µg	23.2 ± 1.0*	41.4 ± 2.7*	54.5 ± 4.6*	59.6 ± 3.6*
	20 µg	31.3 ± 1.0*	44.4 ± 2.7*	61.6 ± 3.6*	71.7 ± 4.4*
β-Citronellol	8.6 µg	28.3 ± 3.6*	47.5 ± 6.6*	60.6 ± 1.7*	67.7 ± 2.0*
	17.2 µg	38.4 ± 1.0*	59.6 ± 3.6*	77.8 ± 2.0*	84.8 ± 1.7*
Fenchol	10 µg	6.1 ± 1.7	17.2 ± 3.6*	30.3 ± 5.2*	36.4 ± 5.2
	20 µg	87.9 ± 6.3*	93.9 ± 4.6 rs	96.0 ± 4.0*	100.0 ± 0.0*
Isomenthol	10 µg	34.3 ± 7.1*	37.4 ± 7.1*	43.4 ± 4.4*	50.5 ± 3.6*
	20 µg	98.0 ± 2.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Linalool	8.6 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	17.2 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Menthol	10 µg	24.2 ± 3.0*	33.3 ± 5.2*	57.6 ± 1.7*	63.6 ± 1.7*
	20 µg	40.4 ± 3.6*	68.7 ± 2.7*	83.8 ± 2.7*	89.9 ± 1.0*
Thymol	10 µg	39.4 ± 6.3*	55.6 ± 6.1*	73.7 ± 6.1*	79.8 ± 4.4*
	20 µg	66.7 ± 1.7*	76.8 ± 2.0*	86.9 ± 2.7*	100.0 ± 0.0*
Carvacrol	9.8 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	19.6 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Nerol	8.8 µg	50.5 ± 2.7*	85.9 ± 4.0*	100.0 ± 0.0 v	100.0 ± 0.0*
	17.6 µg	58.6 ± 7.3*	86.9 ± 3.6*	100.0 ± 0.0 v	100.0 ± 0.0*
Terpinen-4-ol	9.3 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	18.6 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
α-Terpineol	10 µg	25.3 ± 2.7*	33.3 ± 1.7*	62.6 ± 2.7*	72.7 ± 1.7*
	20 µg	97.0 ± 3.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
			Epoxides		
1,8-Cineole	9.2 µg	98.0 ± 8.6*	100.0 ± 0.0*	100 .0± 0.0*	100.0 ± 0.0*
	18.4 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Limonene oxide	9.3 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	18.6 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
			Ketones and aldehydes		
Camphor	10 µg	23.2 ± 5.3*	31.3 ± 5.6*	49.5 ± 3.64*	61.6 ± 2.7*
	20 µg	79.8 ± 4.4*	88.9 ± 2.7*	96.0 ± 3.49*	100.0 ± 0.0*
Carvone	9.6 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	19.2 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Cumin aldehyde	9.8 µg	93.9 ± 1.7*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	19.6 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Dihydrocarvone	9.3 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	18.6 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Fenchone	9.5 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	19.0 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Menthone	9.0 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	18.0 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Esters					
Borneol acetate	9.9 µg	69.7 ± 12.1*	94.9 ± 3.6*	100.0 ± 0.0*	100.0 ± 0.0*
	19.8 µg	98.0 ± 1.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Geraniol acetate	9.2 µg	56.6 ± 1.0*	85.9 ± 1.0*	100.0 ± 0.0*	100.0 ± 0.0*
	18.4 µg	60.0 ± 1.0*	86.9 ± 1.0*	100.0 ± 0.0*	100.0 ± 0.0*
Linalool acetate	9.0 µg	44.4 ± 3.6*	69.7 ± 4.6*	94.9 ± 2.7*	100.0 ± 0.0*
	18.0 µg	44.4 ± 1.6*	73.7 ± 5.6*	99.0 ± 1.0*	100.0 ± 0.0*
Nerol acetate	9.1 µg	36.4 ± 0.0*	50.5 ± 1.0*	84.8 ± 4.6*	92.9 ± 3.6*
	18.2 µg	48.5 ± 1.7*	61.6 ± 2.0*	99.0 ± 1.0*	100.0 ± 0.0*

[a] Mean ± SE of three replicates, each set-up with 20 adults in relation to the time 0.

[b] Exposure time (h). * Statistically different from negative controls (ethanol and sterile water) at $p<0.05$ according to LSD test.

Table 3. Toxic effects of volatile aromatic compounds to adults of *S. granarius* (L.).

Treatments	Dose	Mean mortality (%)[a]			
		24[b]	48[b]	72[b]	96[b]
Anethole	10 µg	46.5 ± 2.7*	66.7 ± 1.7*	86.9 ± 2.7*	100.0 ± 0.0*
	20 µg	61.6 ± 1.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
p-Anisaldehyde	11.2 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	22.4 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Benzyl acetate	10.5 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	21 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Cinnamyl alcohol	10.4 µg	26.3 ± 3.6*	33.3 ± 3.0*	38.4 ± 3.6*	44.4 ± 3.6*
	20.8 µg	56.6 ± 4.4*	61.6 ± 3.6*	76.8 ± 3.6*	83.8 ± 4.0*
Cinnamyl aldehyde	10.5 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
	21 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
Eugenol	10.6 µg	59.6 ± 2.7*	90.9 ± 1.7*	100.0 ± 0.0*	100.0 ± 0.0*
	21.2 µg	74.7 ± 3.6*	94.9 ± 2.7*	100.0 ± 0.0*	100.0 ± 0.0*
Guaiazulene	10 µg	4.0 ± 1.0	46.5 ± 4.4*	64.6 ± 4.4*	68.7 ± 4.4 hıj
	20 µg	11.1 ± 2.0	61.6 ± 2.7*	83.8 ± 3.6*	92.9 ± 2.0 lm
Isoeugenol	10.8 µg	36.4 ± 1.7*	43.4 ± 2.0*	78.8 ± 1.7*	100.0 ± 0.0*
	21.6 µg	49.5 ± 5.6*	66.7 ± 4.6*	100.0 ± 0.0*	100.0 ± 0.0*
Ethyl cinnamate	10.5 µg	1.0 ± 1.0	8.1 ± 1.0	16.2 ± 1.0*	100.0 ± 0.0*
	21 µg	42.4 ± 1.74*	61.6 ± 2.7*	92.9 ± 3.6*	100.0 ± 0.0*
Methyl cinnamate	10 µg	34.3 ± 4.4*	55.6 ± 2.7*	67.7 ± 3.6*	73.7 ± 3.6*
	20 µg	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*	100.0 ± 0.0*
p-Vanillin	10 µg	15.1 ± 5.2	25.3 ± 4.4*	42.4 ± 1.7*	48.5 ± 3.5*
	20 µg	29.3 ± 2.7*	39.4 ± 1.7*	61.6 ± 2.7*	70.7 ± 3.6*

[a] Mean ± SE of three replicates, each set-up with 20 adults in relation to the time 0.

[b] Exposure time (h).

* Statistically different from negative (ethanol and sterile water) control at p<0.05 according to LSD test.

Achieved results and those reported previously clearly indicated the variations in the effects of monoterpenes and aromatic compounds in regard to the stage, the species of insect. Failing that, DDVP which is an effective chemical pesticide was used in this study and 10-20 µL DDVP with a standard of pesticide applications caused 100 % mortality on *S. granarius* after 24 h exposure. Excessive usage of such pesticides causes environmental pollution [77, 78]. Furthermore, it was reported that using of DDVP increased the human cancer risk [79, 80]. For this reason, tested pure compounds proved to be promising as control alternatives against stored product insects such as *S. granarius*. In addition to, there is no statistically (p< 0.01) difference between the 24 h results of DDVP and nine monoterpenes (linalool, terpinen-4-ol, 1,8-cineole, limonene oxide, carvone, cumin aldehyde, dihydrocarvone, fenchone, menthone) and four aromatic compounds (*p*-anisaldehyde, benzyl acetate, cinnamyl aldehyde, methyl cinnamate) and 100% mortality was detected in these compounds at all times and doses. (Tables 1-3 and Figures S2-S5). In the view of these results, it can be considered using green insecticides place of synthetic insecticides which have adverse effects on animals and humans health. The focus over the last few years has been on the determination of the insecticidal activity of isolated chemical compounds from plant extracts in order to find out the most biologically active chemical components [34, 45, 81-84].

Table 4. Durations for 50% and 90% mortality of *S.granarius* (L.) adults the adults of the pest exposed to 10 µL or 10 µg doses of the monoterpenes and aromatic compounds.

		Duration (h)	
	Dose	**50% Mortality**	**90% Mortality**
Monoterpenes hydrocarbons			
Camphene	10 µg	856.5	70337.4
3-Carene	8.7 µg	29.8	77.0
β-Citronellene	7.6 µg	50.6	127.5
Limonene	8.4 µg	32.1	71.7
Myrcene	7.9 µg	40.1	85.2
α-Pinene	8.6 µg	70.1	309.6
β-Pinene	8.7 µg	26.4	63.0
γ –Terpinene	8.5 µg	35.9	121.6
Oxygenated monoterpenes			
Alcohols			
Borneol	10 µg	65.2	386.9
β-Citronellol	8.6 µg	55.8	288.6
Fenchol	10 µg	137.2	584.3
Isomenthol	10 µg	113.8	9754.7
Linalool	8.6 µg	*	*
Menthol	10 µg	63.9	313.3
Thymol	10 µg	35.3	170.5
Carvacrol	9.8 µg	*	*
Nerol	8.8 µg	24.6	47.2
Terpinen-4-ol	9.3 µg	*	*
α-Terpineol	10 µg	55.9	212.4
Epoxides			
1,8-Cineole	9.2 µg	14.0	25.2
Limonene oxide	9.3 µg	*	*
Ketones and aldehydes			
Camphor	10 µg	72.8	400.7
Carvone	9.6 µg	*	*
Cumin aldehyde	9.8 µg	6.6	18.9
Dihydrocarvone	9.3 µg	*	*
Fenchone	9.5 µg	*	*
Menthone	9.0 µg	*	*
Esters			
Bornyl acetate	9.9 µg	18.4	36.6
Geranyl acetate	9.2 µg	22.7	45.6
Linalyl acetate	9.0 µg	28.41	63.2
Neryl acetate	9.1 µg	35.9	97.7
Aromatic compounds			
Anethole	10 µg	28.4	74.4
p-Anisaldehyde	11.2 µg	*	*
Benzyl acetate	10.5 µg	*	*
Cinnamyl alcohol	10.4 µg	156.1	6177.4
Cinnamyl aldehyde	10.5 µg	*	*
Eugenol	10.6 µg	21.6	42.2
Guaiazulene	10 µg	59.9	140.8
Isoeugenol	10.8 µg	37.8	95.6
Ethyl cinnamate	10.5 µg	46.5	169.8
Methyl cinnamate	10 µg	40.4	218.6
p-Vanillin	10 µg	100.7	551.6

*No durations are computed because the ratios of response counts to subject counts are the same, i.e. the slope is zero.

4. Conclusion

The present study demonstrates the possibility of using the test monoterpenes and aromatic compounds such as linalool, carvacrol, terpinen-4-ol, 1,8-cineole, limonene oxide, carvone, cumin aldehyde, dihydrocarvone, fenchone, menthone, bornyl acetate, *p*-anisaldehyde, benzyl acetate, cinnamyl aldehyde and ethyl cinnamate as insecticides against *S. granarius* adults. The effect of these volatile components at low concentrations might reduce the chemical residues and environmental pollution. However, further studies also need to be conducted to evaluate the cost, efficacy and safety of these compounds and essential oils on wide range of pests in commercial store. The diverse activities of the test monoterpenes warrant further research into their potential development as compounds for the control of wheat weevils.

Supporting Information

Supporting Information accompanies this paper on http://www.acgpubs.org/RNP

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