

TOXICITY OF ESSENTIAL OIL OF *AGASTACHE FOENICULUM* (PURSH) KUNTZE TO *ORYZAEPHILUS SURINAMENSIS* L. AND *LASIODERMA SERRICORNE* F.

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Abstract: The essential oil of aerial parts of *Agastache foeniculum* (Lamiaceae) was isolated by hydrodistillation method and constituents of oil were analyzed by gas chromatography mass spectrometry (GC-MS) method. Methyl chavicol, 1,8-cineole, 1-octen-3-ol, 3-octanone and germacrene D the predominant components of oil. Methyl chavicol (94.003%) was identified as a major component in the oil. Essential oil was tested for toxicity against adults of *Oryzaephilus surinamensis* L. and *Lasioderma serricornne* F. The influence of different concentrations of the essential oil vapours on adult mortality was significant. Data of probit analysis showed that a lethal concentration of the essential oil to kill 50% of the population (LC_{50}) for adults of *O. surinamensis* and *L. serricornne* were 18.781 and 21.565 μ l/l respectively. *O. surinamensis* was more susceptible than *L. serricornne* at the exposure time 24 h. The results demonstrated that mortality increased with the increase in concentration and exposure time. These results showed that the essential oil from *A. foeniculum* could be applicable the management of population of stored-product beetle pest.

Key words: essential oil, *Agastache foeniculum*, fumigant toxicity, *Oryzaephilus surinamensis*, *Lasioderma serricornne*

INTRODUCTION

The boom in human population posed a great problem of food scarcity. Stored grain insect pests were damaging our stored agricultural commodities and were responsible for 10–40% of loss worldwide annually (Raja *et al.* 2001). In such a situation, protection of stored grain and agricultural products against insect infestation is an urgent need. Synthetic insecticides and fumigation are the main methods in stored product insect pest's control. Furthermore, an uncontrolled use of these synthetic insecticides causes a great hazard for environment and consumers due to residues (Bell and Wilson 1995; Shaaya *et al.* 1997; Isman 2006). Thus, there is a considerable interest in developing natural products that are relatively less damaging to the mammalian health and environment than existing conventional pesticides, as alternatives to non-selective synthetic pesticides (Jembere *et al.* 1995; Okonkwo and Okoye 1996; Raja *et al.* 2001). Many plant products were evaluated for their toxic properties against different stored grain pests, especially in the form of essential oil (Shaaya *et al.* 1997; Papachristos and Stamopoulos 2002).

O. surinamensis L. (Coleoptera: Silvanidae), is one of the most serious and destructive insect pest of grains stored in bulk condition. This insect feeds on a variety of products including most grains and grain products, dried

fruits, fast foods, nuts, seeds, yeast, sugar, candy, tobacco, dried meat, and in fact, most plant products used as human food (Metcalf and Flint 1979).

The cigarette beetle, *Lasioderma serricornne* F. (Coleoptera: Anobiidae), damages a wide range of stored products including cured tobacco leaves, cigarettes, cocoa beans, cereals, cereal products, oilseeds, spices, dried fruits, and some animal products (Ashworth 1993).

The objective of this study was to analyze chemical constituents of essential oil of *A. foeniculum* (Pursh) Kuntze aerial parts and to identify the active chemical constituents of the essential oil as well as evaluate its fumigant activity of against two major coleopteran pests (*O. surinamensis* and *L. serricornne*) of stored food commodities. The results of this study provide the scientific evidence concerning a possible use of plant oils and their constituents as alternatives to synthetic fumigants for pest control in durable agricultural commodities.

MATERIALS AND METHODS

Insects

O. surinamensis and *L. serricornne* were reared in a glass container (1 l) containing wheat flour that was covered with a fine mesh cloth for ventilation. The cultures were maintained in the dark in an incubator set at $27\pm 2^{\circ}\text{C}$ and

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60±5% RH. Parent adults were obtained from laboratory stock cultures maintained at the Entomology Department, University of Urmia, Iran. Adult insects, 7–14 days old, were used for fumigant toxicity tests. All experimental procedures were carried out under the same environmental conditions as use for the cultures.

Plant material and extraction of essential oil

Aerial parts from 1.5 cm of the top of *A. foeniculum* were collected at the flowering stage from plants grown in the experimental farm at the Department of Horticultural University of Urmia, Urmia, Iran, between April to September 2008. Plant seeds were provided by the University of Budapest, Hungary. The specimen plants were air dried in shade at room temperature (26–28°C) for 14 days. The dried materials were stored in a refrigerator until needed.

The essential oil was isolated from dried plant samples by hydrodistillation method using a Clevenger type apparatus for 4 h. The hydrodistillation of *A. foeniculum* (each time 20 g) gave the oil yield of 1.7%. The oil was dried over anhydrous Na₂SO₄ and stored in a refrigerator at 4°C until required.

Analysis of essential oil

The constituents of *A. foeniculum* essential oil were analyzed by gas chromatography mass spectrometry (GC-MS) (Thermo-UFM). The GS conditions were as follows: capillary column ph-5 (10 m x 0.1 mm, film thickness 0.4 µm); helium as a carrier gas (0.5 ml/min); oven temperature program, initially 60°C rising to 285°C (80°C/min, 3 min); injector and detector temperature of 280°C. The identification of individual compounds was based on comparison of their relative retention times with those of authentic samples on a capillary column, and by matching their mass spectra of peaks with those obtained from authentic samples and published data (Davies 1990).

Fumigant toxicity

Concentrations of 3.57 µl to 42.85 µl and 7.14 µl to 46.42 µl of the oil of *O. surinamensis* and *L. serricornis* respectively, were dissolved in 200 µl acetone and applied to Whatman No. 1 filter paper stripe (4x5 cm), which was dried in air for 2 min. Treated filter paper was placed at the bottom of 280 ml glass jars. Twenty adults (7 to 14 days) of insects were placed in small plastic tubes (3.5 cm diameter and 5 cm height) with open ends covered with cloth mesh. The tubes were hung at the geometrical centre of glass jars, which were then sealed with air-tight lids. Mortality was determined after 24, 48 and 72 h from commencement of the exposure. When no leg or antennal movements were observed, insect was considered dead. Percentage insect mortality was calculated using the Abbott correction formula for natural mortality in untreated control (Abbott 1925).

Statistical analysis

The experiments were arranged using randomized complete block design and the data were subjected to the analysis of variance (ANOVA) using the SAS 8 software. Differences between means were tested with Tukey's test

and values with $p < 0.05$ were considered significantly different. Probit analysis was used to estimate LC₅₀ and LC₉₅ values by SPSS 16.0 software.

RESULTS

Chemical constituents of essential oil

Chemical analysis of essential oil of *A. foeniculum* by means of GC-MS revealed that methyl chavicol, 1,8-cineole, 1-octen-3-ol, 3-octanone and germacrene D were the predominant components of oil. Methyl chavicol (94.003%) and 1,8-cineole (3.334%) were the major constituents (Table 1).

Fumigant toxicity

A. foeniculum oil revealed strong toxicity on adults of *O. surinamensis* and *L. serricornis*. Lethal concentration 50% for the population of insects (LC₅₀) was found for *O. surinamensis* and *L. serricornis* at exposure time 24 h, 18.781 and 21.565 µl/l, respectively (Table 2). With the increase of concentration of oil, increased mortality of the two insects. Furthermore, with the increase of exposure time of insects to the oil, mortality increased and LC₅₀ decreased to 7.853 and 6.240 µl/l at time 72 h for *O. surinamensis* and *L. serricornis*, respectively (Table 2, Fig. 1).

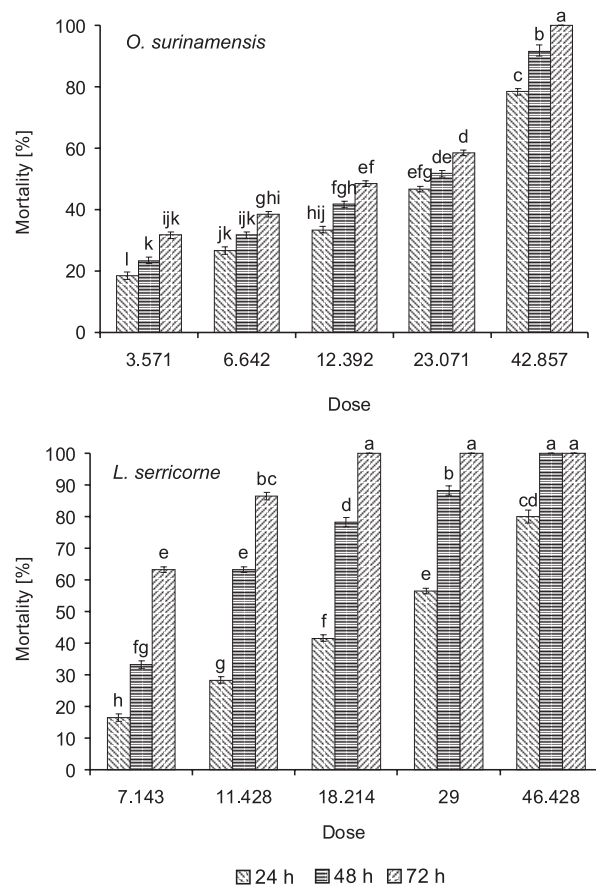


Fig. 1. Mean mortality (%) ±SE of *O. surinamensis* and *L. serricornis* exposed to different doses of essential oil of *A. foeniculum* (µl/l) after 24, 48 and 72 hours. Different letters in the top of columns are significantly different at 5% level

Lethal times for 50% of population of insects (LT₅₀) were 13.463 and 11.601 h for *O. surinamensis* and

Table 1. Chemical constituents of the essential oil from *A. foeniculum*

Row	Component	Retention index	Percentage [%]
1	1-octen-3-ol	977	0.461
2	3-octanone	985	0.407
3	1,8-cineole	1 058	3.334
4	octen-3-yl-acetate	1 108	0.386
5	methyl chavicol	1 200	94.003
6	α -copaene	1 375	0.029
7	β -boarbone	1 386	0.084
8	E-caryophyllene	1 418	0.058
9	germacrene D	1 485	0.430
10	bicyclgermacrene	1 500	0.020
11	Spathulenol	1 570	0.039
12	α -maurolool	1 643	0.014
13	β -eudesmol	1 650	0.015
14	dihydro eudesmol	1 664	0.013

Table 2. LC_{50} and LC_{95} calculated for mortality within 3 days of exposure of *O. surinamensis* and *L. serricorne* to fumigation with essential oil of *A. foeniculum*

Insect	Time [h]	LC_{50} [μ l/l]	LC_{95} [μ l/l]	Intercept [a]	Slop [b]	χ^2 [DF = 3]
<i>O. surinamensis</i>	24	18.781	261.353	3.103	1.490	4.812*
	48	11.312	139.767	3.128	1.777	4.977*
	72	7.853	95.824	3.635	1.525	2.956*
<i>L. serricorne</i>	24	21.565	126.539	2.145	2.140	1.132*
	48	9.651	34.382	2.065	2.981	3.251*
	72	6.240	13.569	1.127	4.871	1.581*

*significant difference at $p < 0.05$

Table 3. LT_{50} and LT_{95} calculated for mortality within 3 days of exposure of *O. surinamensis* and *L. serricorne* to fumigation with *A. foeniculum* oil at the highest dose (42.857 μ l/l for *O. surinamensis* and 46.428 μ l/l for *L. serricorne*)

Insect	LT_{50} [h]	LT_{95} [h]	Intercept [a]	Slop [b]	χ^2 [DF = 1]
<i>O. surinamensis</i>	13.463	49.000	1.690	2.932	2.189*
<i>L. serricorne</i>	11.601	55.295	2.418	2.425	4.171*

*significant difference at $p < 0.05$

L. serricorne at the highest concentration, respectively (Table 3). In all cases, considerable differences in mortality of insects to essential oil vapour were observed with different concentrations and times (Table 2, Fig. 1). On the other hand, the increase of susceptibility of the two insect pests associated with the increase of concentrations of oil and time of exposure.

Probit analysis showed that *O. surinamensis* was more susceptible ($LC_{50} = 18.781 \mu$ l/l) to *A. foeniculum* oil than *L. serricorne* ($LC_{50} = 21.565 \mu$ l/l) at the exposure time 24 h. As the exposure time increased to 72 h, susceptibility of *L. serricorne* increased and LC_{50} achieved the value of 6.240 μ l/l. In the contrary, until this time (time 72 h) susceptibility of *O. surinamensis* was little increased ($LC_{50} = 7.853 \mu$ l/l).

DISCUSSION

In all cases, this study demonstrated strong toxicity of *A. foeniculum* oil to *O. surinamensis* and *L. serricorne* that is a major pest of stored products. The insecticidal activity varied with insect species, different concentrations of the oil and exposure time. The results showed a higher susceptibility in *O. surinamensis* than in *L. serricorne* at the time 24 h and a higher susceptibility in *L. serricorne* than in *O. surinamensis* at the time of 72 h. The increased exposure time to 72 h, also increased susceptibility of the two insect pests to essential oil and aggravated its mortality.

Jacobson (1989) demonstrated that the most promising botanical insect-control agents are in the families: Annonaceae, Asteraceae, Canellaceae, Lamiaceae, Meliaceae, and Rutaceae. *A. foeniculum* is a perennial member in plants family Lamiaceae. In spite that essential oil of

the species of *A. rugosa* was evaluated for insecticidal and nematocidal activity (Kim *et al.* 2003; Choi *et al.* 2007), toxicity of *A. foeniculum* oil not investigated against insect pests until now.

Toxic effects observed for *A. foeniculum* oil on the insect species tested suggested that this action could be attributed to the oil main components. Charles *et al.* (1991), investigated characteristics of some of the essential oils from *Agastache* genus and found out that methylchavicol was a major constituent of *A. foeniculum* oil and this was also proved in this study. Toxicity of methylchavicol was not detected against insect pest of a stored product, but 1,8-cineole (another major constituent of *A. foeniculum* oil) was reported as a toxic agent on some insects (Lee *et al.* 2004; Erler 2005).

The development of natural or biological insecticides would help to decrease the negative effects (residues, resistance and environmental pollution) of synthetic insecticides. With this respect, bio-insecticides may be not involved in resistance of the pest and may be less toxic to environment. In the present study, essential oil of *A. foeniculum* was the most toxic against *O. surinamensis* and *L. serricornis* adults. The essential oil of *A. foeniculum* used as medicine is considered to be less harmful than most conventional insecticides. Consequently, the possibility of employing these natural fumigants to control insects in stored products may need further investigation. However, further studies need to be also conducted to evaluate the cost and efficacy of these essential oils on a wide range of insect pests. The mode of action of essential oils is yet to be confirmed, but it appears that death of the insects may be due to suffocation and inhibition of different biosynthetic processes of the insect's metabolism (Don-Perdo 1989).

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POLISH SUMMARY

TOKSYCZNOŚĆ OLEJKÓW ETERYCZNYCH AGASTACHE FOENICULUM (PURSH) KUNZE DLA ORYZAEPHILUS SURINAMENSIS L. I LASIODERMA SERRICORNE F.

Izolowano olejek eteryczny z części nadziemnych *Agastache foeniculum* (Lamiaceae), wykorzystując metodę hydrodestylacji, a jego składniki izolowano przy wykorzystaniu metody spektrometrii masy (GC-MS). Prze-

ważącymi składnikami olejku były: chawikol metylu, 1,8-cineol, 1-octen-3-ol, aceton i germacrene D. Chawikol metylu (94,003%) był określony jako główny komponent olejku. Testowano jego toksyczność w stosunku do dorosłych osobników *Oryzaephilus surinamensis* i *Lasioderma serricorne*. Wpływ różnych stężeń par olejku na śmiertelność dorosłych osobników był istotny. Analiza żywienia wykazała, że zabójcze stężenie olejku eterycznego potrzebne do zabicia 50% populacji (LC_{50}) dla osobników dorosłych

O. surinamensis i *L. serricorne* wynosiło odpowiednio 18,781 i 21,565 $\mu\text{l/l}$. *O. surinamensis* była bardziej wrażliwa niż *L. serricorne* przy czasie ekspozycji 24 godziny. Wyniki wykazały, że śmiertelność wzrastała wraz ze wzrostem stężenia oraz czasu ekspozycji. Wykazano, że olejek eteryczny z *A. foeniculum* mógłby być stosowany w regulowaniu populacji szkodliwego, dla przechowywania produktów, chrząszcza.