EFFECTS OF ESSENTIAL OIL FORMULATIONS ON THE ADULT INSECT *TRIBOLIUM CASTANEUM* (HERBST) (COL., TENEBRIONIDAE)

EFIKASNOST ETARSKIH ULJA NA ODRASLE INSEKTE *TRIBOLIUM CASTANEUM* (HERBST) (COL., TENEBRIONIDAE)

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ABSTRACT

Stored product pests such as *Tribolium castaneum* (Herbst, 1979) are a major problem. Adult insects were obtained from laboratory cultures maintained in the dark in incubators at 25± 1C and 70–80% r.h., reared on wheat flour and fed with flour disks containing a known concentration of essential oil of 9 plants. The chemical components of essential oil of 3 plants, collected on the area of Montenegro, were also identified using GC-MS analysis. The results of insecticidal effect of essential oils were discussed. Also, mortality rate of adult insects was tested. In this research, the essential oils of *C. glandulosa* which were rich in monoterpene alcohols carvacrol and contained ketonic component showed strong insecticidal and fumigant activity against adults of *T. castaneum*. Less toxic effect showed essential oils of *Satureja montana* which had a lower carvacrol and ketonic content. On the other hand, essential oils of *Teucrium polium* which did not contain ketonic component did not show any activity. Therefore, it was observed that essential oils of *C. glandulosa* with concentration of 1.14% showed powerful toxic and repellent effect, with very high mortality rate after 24h (56,67%).

Keywords: *Calamintha glandulosa,* essential oils, mortality rate, toxicity,*Tribolium castaneum*

REZIME

Štetočine skladišta, kao što je *Tribolium castaneum* (Herbst, 1979) predstavljaju značajan problem. Odrasli insekti, korišćeni u ovom istraživanju, uzgajani su u laboratoriji u kontrolisanim uslovima (25±1C i 70-80% vlage) na pšeničnom brašnu, a kao hrana korišćeni su diskovi pravljeni od vode i pšeničnog brašna, na koje je nanošena poznata koncentracija eteričnog ulja 9 različitih biljaka. Pomoću GC-MS analize utvđen je sadržaj etarskih ulja 3 biljke sakupljene na području Crne Gore, a to su sledeće: *Calamintha glandulosa, Teucrium polium* i *Satureja montana* . Razmatrano je insekticidno dejstvo etarskih ulja ispitivanih biljaka, takođe i stopa smrtnosti odraslih insekata. U ovom istraživanju, etarsko ulje biljke *Calamintha glandulosa,* pokazuje visok sadržaj monoterpena, odnosno karvakrola i ketonskih komponenti, koje zajedno imaju jak insekticidni i fumigantni efekat na adulte *T. castaneum*. Slabija toksičnost je utvrđena kod etarskog ulja biljke *Satureja montana,* koje je sadžalo nizak nivo karvakrola, kao i ketona. Takođe, etarsko ulje biljke *Teucrium polium,* u kojem nije utvrđen sadržaj ketonskih komponenti nije pokazalo insekticidnu aktivnost. Dakle, uočeno je da etarsko ulje *Calamintha glandulosa* pri koncentraciji od 1.14% pokazuje značajan toksičan i repelentan efekat, sa veoma visokom stopom smrtnosti (56.67%), već nakon 24 sata.

Ključne reči: *Calamintha glandulosa,* etarska ulja, stopa smrtnosti, toksičnost, *Tribolium castaneum*

DETALJAN REZIME

Štetočine skladišta, kao što je *Tribolium castaneum* (Herbst, 1979) predstavljaju značajan problem. Odrasli insekti, korišćeni u ovom istraživanju, uzgajani su u laboratoriji u kontrolisanim uslovima (25±1C i 70-80% vlage) na pšeničnom brašnu. Ogled je postavljen, u petri pusude, u tri ponavljanja za svaku biljku i kontrolu. Postavljeno je 10 odraslih insekata u svaku petri posudu, a kao hrana za insekte korišćeni su diskovi od testa (po jedan disk u svaku posudu) pravljeni mešanjem vode i pšeničnog brašna, na koje je nanošena poznata koncentracija eteričnih ulja 9 različitih biljaka i n-helsan kao kontrola. Biljke su sakupljane na području Srbije i Crne Gore, tokom maja, juna i avgusta meseca. Primenjene su dve koncentracije etarskih ulja: 1.14% i 0.02 %. Pomoću Hewlett-Packard 5973-6890 GC-MS analize utvđen je sadržaj etarskih ulja 3 biljke sakupljene na području Crne Gore, a to su sledeće: *Calamintha*

glandulosa, Teucrium polium i *Satureja montana*. Razmatrano je insekticidno dejstvo etarskih ulja ispitivanih biljaka, takođe i stopa smrtnosti odraslih insekata. Dakle, uočeno je da etarsko ulje *C. glandulosa* pri koncentraciji od 1.14% pokazuje značajan toksičan i repelentan efekat, sa veoma visokom stopom smrtnosti (56.67%), već nakon 24 sata, dok je posle 48h iznosila 83.33%. Stopa smrtnosti u kontroli nije razmatrana, jer je iznosila 0%, odnosno svi insekti su preživeli. U slučaju biljke *Thymus serpyllum* primećena je veoma niska stopa smrtnosti pri istoj koncentraciji od 1.14%, svega 13.33%, posle 48h i 96h. Smrtnost insekata pod dejstvom eteričnih ulja biljaka *Origanum vulgare* i *Teucrium chamaedrys*, nakon 48h iznosila je svega 6.66%, a nakon 96h bila je 10%, što pokazuje veoma slab toksičan efekat. Niža koncentracija od 0.02% etarskih ulja 9 ispitivanih biljaka nije uzrokovala uginuće insekata nakon 24h, 48h i 96h.
U ovom istraživanju, etarsko ulje biljke *C. glandulosa,* koje sadrži 79.91 % karvakrola, oko 14% monoterpenskih ugljovodonika, kao i 3 % ketona zajedno imaju jak insekticidni i fumigantni efekat na adulte *T. castaneum*. Slabija toksičnost je utvrđena kod etarskog ulja biljke S*atureja montana,* koje je sadžalo nizak nivo karvakrola (50.1%), kao i ketona (1.18%). Takođe, etarsko ulje biljke *T. polium,* u kojem nije utvrđen sadržaj ketonskih komponenti nije pokazalo insekticidnu aktivnost. Etarsko ulje *C. glandulosa* koje je sdržalo najveći procenat karvakrola pokazuje i najveću fumigantnu i repelentnu aktivnost. Karvakrola predstavlja samo jednu od mnogobrojnih sastojaka etarskih ulja, koji poseduje biocidni učinak na insekte. S obzirom na rezultate naših istraživanja, korišćenje prirodnih supstanci, kao šuo su etarska ulja, predstavljaj dobru alternativnu metodu u kontroli insekata.

INTRODUCTION

Tribolium castaneum (Herbst,1797) (Coleoptera: Tenebrionidae) known as rust red flour beetle, are among the most important stored-grain pests. *T. castaneum* is one of the key pests of stored products and stored grains throughout the world (Sinha and Watters, 1985). It is one of the most common insect pests worldwide of flour mills, grocery shops, and warehouses (Garcìa et al., 2005, Jemâa, J.M.B et al., 2012). Essential oils are believed to act as allelopathic agents or as irritants that protect plants from predation by insects and infestation by parasites (Simpson, 1995). Essential oils and their constituents have also been shown to be a potent source of botanical pesticides (Singh and Upadhyay, 1993). The essential oils of several spices like anise (*Pimpinella anisum* L.) and peppermint (*Mentha piperita* L.) have been found to have fumigant toxicity to four major stored product pests, *R. dominica, Tribolium castaneum* (Herbst), *S. oryzae* and *Orzyaephilus surinamensis* (L.) (Shaaya et al., 1991).Ho et al. (1996) found that the essential oils of garlic are insecticidal to *T. castaneum* and *Sitophilus zeamais* Motschulsky. The essential oils of nutmeg seeds (*Myristica fragrans* Houtt) (Huang et al., 1997) and cinnamon bark (*Cinnamomum aromaticum* Nees)

(Huang and Ho, 1998) are also toxic to these two species of insects (Huang et al., 2000).

Also, repeated applications of liquid and gaseous insecticides has disrupted natural enemies' biological control system and led to out breaks of insect pests, wide spread development of resistance, undesirable effects on non-target organisms, and environmental and human health concerns (Subramanyam and Hagstrum,1995; White and Leesch, 1995).
Because of these problems, plants may provide potential alternatives to currently used insect-control agents because plants constitution included a rich source of bioactive chemicals (Wink, 1993).

Much effort has, therefore, been focused on plant-derived materials for potentially useful products as commercial insect-control agents. Little work has been done to manage stored-product insects by using aromatic medicinal plants, despite their excellent pharmacological actions (Tang and Eisenbrand, 1992; Namba, 1993; Kim, 1996). In fact, management of stored product pests, using substances of natural origin, is nowadays the subject of much research (Derbalah and Ahmed, 2011)
Results of insecticidal effect of essential oils, plant-growing on the territory of Serbia and Montenegro, are tested and discussed in this paper.

MATERIALS AND METHODS

Experimental conditions

The experiments on the *Tribolium castaneum* (Herbst, 1797) carried out at the Faculty of Agriculture, University of Novi Sad (Republic of Serbia). Adults *T. castaneum* were obtained from laboratory cultures maintained in the dark in incubators at 25± 1C and 70–80% r.h. *Tribolium castaneum* was reared on wheat flour.

Plant materials

The following aromatic species (Lamiaceae) collected in Serbia and Montenegro were used: *Calamintha glandulosa* L. (*C. glandulosa*), *Satureja Montana* L. (*S. montana*), *Teucrium polium* L. (*T. polium*) , *Salvia officinalis* L. (*S. officinalis*) growing wild at localities near the Adriatic coast in Montenegro; *Thymus serpyllum* L. (*T. serpyllum*), *Thymus marschalianus* L. (*T. marschalianus*), *Teucrium chamaedrys* L. (*T. chamaedrys*), *Origanum vulgare* L. (*O. vulgare*) growing wild in Vojvodina province (northern Serbia) and *Salvia sclarea* L. (*S. sclarea*) growing wild in the south of Serbia.

The aerial parts of flowering plants *T. serpyllum*, *T. marschalianus*, *T. chamaedrys*, *S. montana* and *S.officinalis* were collected in May of 2010; *C. glandulosa* and *T. polium* in June of 2010; *O. vulgare* and *S. sclarea* in August of 2010. Voucher specimens of collected plants were confirmed and deposited at the Herbarium of The Department of Biology, Faculty of Natural Sciences, University of Novi Sad.

Out of 9 tested plants, the essential oils analysis was carried out from 3 plants collected in Montenegro: *C. glandulosa, S. Montana, T. polium*.

Toxicity test

The bioassays were carried out using groups of 10 *Tribolium castaneum*, kept in the petri dishes with transparent covers, fed with flour disks containing a known concentration (1.14%, 0,02%) of essential oils. The flour disks with n-hexane and 10 insects for eachwere used as controls. The experiment was set in three replicates for each plant and control. A no-choice method in which control and treated disks were placed individually in petri dishes, was adopted in this experiment because stored-product insects, often broke the flour disks into small pieces and in the choice method, it is very difficult to determine whether the small pieces come from the control or treated disks.

All the insects were starved for 24 h before use. The disks were placed in petri dishes for weighing, than insects added – the group of 10 in each petri dish. The experimental set-up was left in the incubator. The flour disks and live insects were then weighed again with any mortality of insects recorded. Mortality was checked after 24, 48 and 96 h.

Also petri dishes containing treated flour disks but without the insects were prepared to determine if any decrease in weight had occurred due to evaporation of n-hexane or plant extracts.

Essential Oils Isolation

The air-dried plant materials (30g) were submitted to hydro distillation using a Clevenger-type apparatus for 3 h. The oils were dried over anhydrous sodium sulphate and stored in a sealed vial at low temperature before analysis. The oils were analyzed by GC-MS analysis.

Essential Oils Analysis

Qualitative and quantitative analyses of the essential oils were carried out using a Hewlett-Packard 5973-6890 GC-MS system, operating in EI mode at 70 eV, equipped with a split less injector (200°C) and a flame ionization detector (250°C). Helium was used as carrier gas (1 mL/min), and the capillary column was a HP 5MS (30 m _ 0.25

mm; film thickness, 0.25 ím). The temperature programs were 60-280°C at a rate of 3°C/min and 60- 260°C at a rate of 3°C/min, respectively; split ratio, 1:10. Coelution and MS analysis were based on the identification of individual compounds in comparison to their relative retention times with those of authentic samples (Carl Roth GmbH, Karlsruhe, Germany). For the components, mostly sesquiterpenes and aliphatic compounds, for which reference substances were not available, the identification was performed by matching their retention indices (RIs) and mass spectra with those obtained from authentic samples and/or the NIST/NBS and Wiley libraries spectra as well as with literature data (Adams, 2007).

Data analysis

One-way analysis of variance (ANOVA) of mortality was used to determine the statistical significance compared with the controls; $P<0.001$ were considered significant. The means were separated by Duncan's multiple range test.

RESULTS AND DISCUSSION

Essential Oils Composition

The composition of the essential oils tested can be seen in table (Table 1). The *C. glandulosa* oil contained 79.91% carvacrol, about 14% of monoterpene hydrocarbons and 3% of ketones. The monoterpenehydrocarbons fraction contained p-cymene (4.41%) and γ- terpinene (4.06%). The ketonic component contained pulegone (1.24%), iso-menthone (1.33%) and borneol (0.29%).

The oil from *S.montana* had a lower carvacrol (50.1%) and ketonic (1.18%) content and higher concentrations of monoterpene (over 30%) and sesquiterpene (over 13%) hydrocarbons. The monoterpene hydrocarbon components had a relatively high content of p-cymene (10.12%) and γ-terpinene (8.79%). Sesquiterpene hydrocarbons fraction consisted mostly of caryophyllene (8.71%). The ketonic component contained borneol (1.18%).

The main constituents of *T. polium* oils were phenolic monoterpens carvacrol (67.84%). The second largest component of the oil was sabinen (10.7%) representing almost the whole monoterpene hydrocarbon fraction. The sesquiterpene hydrocarbons fraction contained germacrene D (2.73%), β-bisabolene (2.55%) and caryophyllene (2.34%).

Table 1. Essential oil components

Tabela 1. Komponente etarskih ulja

Components	*C. glandulosa* (%w/w)	*S. Montana* (%w/w)	*T. polium* (%w/w)
α-Thujene	0.52	0.43	0.84
α-Pinene	0.64	0.66	3.65
Sabinene	nd	nd	10.70
1-Octen-3-ol	0.99	0.92	nd
β-Pinene	nd	nd	2.91
β-Myrcene	1.34	1.21	1.56
α-Terpinene	1.05	1.45	nd
***p*-Cymene**	4.41	10.12	0.92
Limonene + β-Phellandrene	0.33	nd	nd
Z-β-Ocimene	nd	2.81	nd
E-β-Ocimene	nd	0.44	nd
γ-Terpinene	4.06	8.79	0.87
(monoterpene)	nd	2.24	nd
iso-Menthone	1.33	nd	nd
Menthol	0.42	nd	nd
Borneol	0.29	1.18	nd
4-Terpineol	0.56	0.60	nd
Carvacrol/thymol, methyl ether	nd	0.61	nd
Pulegone	1.24	nd	nd
Geraniol	nd	1.62	nd
Thymol	0.20	3.58	nd
Carvacrol	79.91	50.1	67.84
Caryophyllene	0.63	8.71	2.34
Z-β-Farnesene	nd	nd	0.82
α-Humulene (α-Caryophyllene)	0.00	1.00	nd
α-Amorphene	0.00	0.41	nd
Germacrene D	nd	nd	2.73
β-Bisabolene	1.33	nd	2.55
γ-Cadinene	0.17	0.33	nd
δ-Cadinene	0.41	0.92	1.10
(sesquiterpene)	nd	0.92	nd
(sesquiterpene)	nd	0.94	nd
α-Cadinol	nd	nd	1.19

nd, not detected

Toxicity on adult

The essential oil of *C.glandulosa* at the highest concentration tested caused the death of 83.33%of the insects after 48h. Table 2 shows that mortality rates in relation to the controls. Mortality rate was not observed in the petri dishes with control and it was zero percent.

When 1.14% concentration of *C. glandulosa* oils was used, 56.67% of the insects died after 24h. Essental oils of other eight examined plants showed no mortality towards the adult insects. After 48h, the most effective formulation was *C.glandulosa* with 83.33 % mortality. After 96h, the oils of previously mentioned plants, showed greatest toxicity (96.67% mortality). In the case of *Thymus serpyllum* lower mortality was observed in the same concentration (1.14%), and the percentage of died insects was 13.33% after 48h and 96h. After 48h, the mortality rate of *Origanum vulgare* and *Teucrium chameadrys* was very low 6.66%. Also, essential oils of these two plants produced a weak toxic effect, and the mortality rate was 10%, after 96h. However, the formulations containing the essential oils of remaining plants produced zero mortality after 48h and 96h.

The lower concentration of essential oils of all nine plants examined was 0.02%, but there was no mortality after 24h, 48h, and 96h.

Table 2. Mortality of *T. castaneum* adult fed for 4 days with formulations containing a known concentration (1.14%, 0.02%) of essential oils.

Tabela 2. Smrtnost odraslih insekata *T. castaneum* hranjenih 4 dana na poznatim koncentracijama (1.14%, 0.02%) etarskih ulja.

Plants	Essential oil (%)	Mortality (%)[2]		
		Time (24h	48h	96h)
Controla		**0.0**	**0.0**	**0.0**
Origanum vulgare	**1.14**	0.00	**6.66b**	**10.00b**
	0.02	0.00	0.00	0.00
Calamintha glandulosa	**1.14**	**56.67a**	**83.33a**	**96.67a**
	0.02	0.00	0.00	0.00
Teucrium polium	**1.14**	0.00	0.00	0.00
	0.02	0.00	0.00	0.00

Thymus marschalianus	**1.14**	0.00	0.00	0.00
	0.02	0.00	0.00	0.00
Salvia officinalis	**1.14**	0.00	0.00	0.00
	0.02	0.00	0.00	0.00
Satureja montana	**1.14**	0.00	0.00	0.00
	0.02	0.00	0.00	0.00
Salvia sclarea	**1.14**	0.00	0.00	0.00
	0.02	0.00	0.00	0.00
Teucrium chamaedrys	**1.14**	0.00	**6.66b**	**10.00b**
	0.02	0.00	0.00	0.00
Thymus serpyllum	**1.14**	0.00	**13.33b**	**13.33b**
	0.02	0.00	0.00	0.00

[2] means within a column of each treatment followed by the same letter are not significantly different (P< 0.001, Duncan's multiple range test)

The essential oils of *C.glandulosa* which was rich in monoterpene alcohols carvacrol and contained ketonic component showed strong insecticidal and fumigant activity against *T. castaneum* adults. Less toxic effect showed essential oils of *S. montana* which had a lower carvacrol and ketonic content. On the other hand, the essential oils of *T. polium* which did not contain ketonic component did not show any activity. The main constituent of *C. glandulosa*, *S. montana* and *T. polium* oils was monoterpene carvacrol alcohol.

Carvacrol has broad insecticidal and acaricidal activity against agriculture, stored-product and medical pests, and it acts as a fumigant (Ayvaz et al., 2010). Also, It is interesting to point out the results of earlier tests of insecticidal carvacrol activity. The insecticidal carvacrol activity was confirmed on the wine fly, *Drosophila melanogaster*. Therefore, it is found that carvacrol has a stronger insecticidal activity than thymol, and that in combination with thymol its insecticidal potential decreases. This indicates the antagonism of the two phenols (Karpouhtsis et al., 1998) when used as an insecticide.

It was also confirmed that carvacrol has insecticidal activity on the brown cockroach, *Blattella germanica*, with highly toxic effect to all forms of its development in contact and topical application as well as a fumigant (Phillips, 2009, Phillips and Appel, 2010, Phillips et al., 2010).

Taking into account our result, it is obvious that the insecticidal activity of the essential oils of the three aromatic plants is not dependent only on this constituent. Karpouhtsis et al. (1998) reported that essential oils of *O. vulgare* and *C. capitatus* with an extremely high content of carvacrol exhibit less insecticidal activity than that of *S. thymbra* that contains a very small amount of this phenol. It could indicate that either other constituents of the aromatic plants essential oils are responsible for their toxicity, or that synergistic and/or antagonistic phenomena exist, and it alters the toxicity of the whole essential oils (Karpouhtsis et al., 1998).

Biosynthetic precursors of thymol and carvacrol are two monoterpenes*γ*-terpinene and *p*-cymene (Karpouhtsis et al., 1998). Thymol is a monoterpenoid and exhibits antibacterial, antifeedant and insecticidal activity (Waliwitiya et al., 2009; Jack IR et al., 2006). The sum of these precursors is higher in the *C. glandulosa* and *S. montana* oils, while in *T. polium* this sum is lower.

Batish et al. (2008) reported that pesticidal activity of eucalyptus oils has been due to the components such as α-terpineol, p-cymene, γ-terpinene and limonene (Batish et al., 2008). The sum of these components is higher in the *C. glandulosa* and *S. montana* oils than in *T. polium*, respectively.

The poor toxic effect of *T. polium* could be explained by the lower *γ*-terpinene,*p*-cymene, α-terpineol, p-cymene, γ-terpinene and limonene content of its oils. Also, the major toxicity of *C. glandulosa* could be explained by the activity of components which were found only in these oils: izo-menton, pulegon, mentol and limonen. Lognay et al. (2002) reported that the essential oil of *A. scrophulariaefolia* L. was shown a fumigant effect against the stored grain insect pest – *Callosobruchus maculatus*. The main constituents identified were limonene (11.4%), menthone (7.6%), isomenthone (49.7%) andpulegone (19.8%) (Lognay et al., 2002).

The less powerful toxic effect of *S. montana* compared to *C. glandulosa* could be explained by the lower carvacrol and ketonic content of its essential oils.

CONCLUSION

Essential oils from different plant species and their active components are natural sources of biocides. Carvacrol is just one of examples of active components of essential oils of many plants that possess insecticide and acaricide effect. Therefore, it was observed that essential oils of *C. glandulosa* with concentration of 1.14% showed powerful toxic and repellent effect, with very high mortality rate after 24h (56.67%). Our results show that the use of natural substances such as essential oil, could be an alternative method of insect control.

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