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Research Article
Biological Activities of *Hypericum perforatum* L. Essential Oil Against Red Flour Beetle, *Tribolium castaneum* (Herbst) (Coleoptera: Tenebrionidae)

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Abstract

Background: Exploration of eco-friendly pesticides is necessary because of the side effects of synthetic chemicals such as environmental hazards, pest resistance and toxic effects on non-target organisms. Many plant essential oils have indicated potential insecticidal effects against several insect pests. **Materials and Methods:** The essential oil of *Hypericum perforatum* L. (Hypericaceae) was obtained by steam distillation and its chemical composition along with fumigant toxicity and antifeedant activity were evaluated against the adults of red flour beetle, *Tribolium castaneum* (Herbst). **Results:** Fourteen components were identified in the essential oil that characterized by the following main constituents: Decane (59.58%), dodecane (12.93%), ethylcyclohexane (6.84%), 5-methylnonane (4.71%), 3-methylnonane (4.32%) and tetradecane (3.82%). Results of fumigant toxicity showed *H. perforatum* essential oil has remarkable biological activity which depended on essential oil concentrations and exposure times. As the exposure time increased the mortality rate increased and the LC_{50} values decreased from 15.048 μL L^{-1} air at 24 h to 11.743 μL L^{-1} air at 72 h exposure time. About 24 h sub-lethal concentrations (LC_{25}-LC_{45} μL L^{-1} air) were used in antifeedant bioassays. Essential oil of *H. perforatum* had significant antifeedant effects on *T. castaneum*. The LC_{45} (14.13 μL L^{-1} air) caused highest Feeding Deterrence Index (FDI) of 95.78%. **Conclusion:** Results of present study revealed that essential oil of *H. perforatum* have potential insecticidal effects on *T. castaneum* and this essential oil can be a natural alternative to harmful chemical insecticides in the integrated management of *T. castaneum*.

Key words: Essential oil, feeding deterrence index, fumigant toxicity, *Hypericum perforatum*, *Tribolium castaneum*

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Darkling beetles (Coleoptera: Tenebrionidae) are a large group of insects comprising more than 10,000 species. Among darkling beetles, *Tribolium* spp., are famous for producing toxic quinines which contaminate flour products (Gorham, 1991). Red flour beetle, *Tribolium castaneum* (Herbst) (Coleoptera: Tenebrionidae), reduces the quantity and quality of grain-based products and can be a major pest in stored products. It has been found in a wide range of dried materials of animal and plant origin, especially beans, dried fruits, flour, grains, nuts, peas and spices (Rees, 2007; Caballero-Gallardo *et al*., 2011; Khan *et al*., 2014). Synthetic fumigants are the most effective tools for protection of stored commodities against insect pests. However, several issues have been discussed in the excessive use of these chemicals, such as adverse effects on the environment, toxicity to humans, the reduction of the populations of natural enemies, secondary pest's outbreaks and development of resistance in insect populations (Bughio and Wilkins, 2004; Collins *et al*., 2005). Therefore, these problems have highlighted the need for development of safe and natural alternatives to synthetic ones.

Botanical pesticides especially plant essential oils have the advantage of safe and selective materials and novel mode of action against pests that can reduce chemical residue and pest's resistance (Isman, 2006; Ebadollahi and Jalali Sendi, 2015). A commonly used method for commercial production of essential oils is steam distillation (Burt, 2004) and because of the mode of extraction, the essential oils with volatile molecules such as terpenes and aromatic components may be used as natural fumigants (Bakkali *et al*., 2008). In some countries, essential oils isolated from aromatic plants are used customarily and commercially to protect grains against storage pests (Regnault-Roger *et al*., 2012; Isman and Grieneisen, 2014).

Hypericum spp., from Hypericaceae family includes nearly 484 species (Guedes *et al*., 2012). Many of them have an extended customary importance as medicinal aromatic plants (Rabanal *et al*., 2002; Camejo-Rodrigues *et al*., 2003; Ferreira *et al*., 2006). *Hypericum perforatum* L., (common St John's wort, Tipton's weed, rosin rose or Klamath weed) has been included in the pharmacopeia of many countries (Ivanova *et al*., 2005; Jaric *et al*., 2007). The preparative formulations of *H. perforatum* are sold for the treatment of mild to moderate depression in the USA and Europe (Ahmed *et al*., 2013). The insecticidal effects of essential oils from some *Hypericum* spp., have been assessed in previous studies (Kordali *et al*., 2012; Rouis *et al*., 2013) and these plant materials have been candidates for future work in the management of pests. Therefore, the aims of present study are to evaluate the toxicity and antifeedant effects of the essential oil extracted from *H. perforatum* against *T. castaneum*. In addition, the chemical composition of this essential oil was determined by gas chromatography-mass spectrometry (GC-MS) analyses.

MATERIALS AND METHODS

Plant material and essential oil extraction: Aerial parts from 4 cm of the top of *H. perforatum* were collected at flowering stage from Heyran region (38°23′40.92″N, 48°36′3.96″E), Ardabil province, Iran. After its identification, the specimens were air-dried in the shade at room temperature (26-28°C) for 14 days and the essential oil was extracted from dried plant samples by hydro-distillation method with a Clevenger type apparatus. Extraction conditions were: 100 g of air dried sample, 1:10 plant material/water volume ratio, 3 h distillation. Anhydrous sodium sulfate was used to eliminate water after extraction. Extracted essential oil was kept in a refrigerator at 4°C.

Analysis of the essential oil: The GC-MS analysis was carried out on a Hewlett-Packard (HP, Palo Alto, CA, USA) HP 7890A GC armed with a split/split less injector and 5975C mass discerning detector system. Chromatographic separation was carried out in a HP-5 capillary column (30×0.25 mm, 0.25 µm in thickness). The MS was operated in the EI mode (70 eV). The GC-MS interface, ion source and quadruple temperatures were set at 280, 230 and 150°C, respectively. The injector temperature was set at 250°C, the column temperature program started at 50°C for 3 min, increased by 10°C min^{-1} to 110°C and by 10°C min^{-1} to 180°C and was maintained for 2 min. Helium (99.999%) was used as the carrier gas with a flow rate of 1 mL min^{-1}. Identification of spectra was carried out by comparing their retention times, mass spectra fragmentation with those on the stored Wiley 7n.1 mass computer library and NIST (National Institute of Standards and Technology) (Adams, 2007).

Insect rearing: The *T. castaneum* obtained from a colony housed at the Department of Plant Protection, University of Tehran, Iran. The insects were reared in plastic containers (20 cm length×14 cm width×9 cm height) containing a wheat flour and yeast mixture (10:1 w/w). The top of the containers were covered with a fine mesh cloth for aeration and to prevent the beetles from escaping. Insects were kept in

darkness at 27±1°C and 60±5% RH. The insects used for these experiments were 1-7 days old adults.

Fumigant toxicity: The fumigant toxicity was determined according to the method of Negahban *et al.* (2007). Concentrations of 9-20 µL L^{-1} air were dissolved in 200 µL acetone and applied to filter paper strips (3×4 cm, Whatman No. 1), which were air-dried for 5 min. Treated filter papers were placed at the bottom of 1 L glass jars. For prevention of contact between the essential oil and the insects, 20 adult insects were placed in small plastic tubes (3.5 cm diameter and 5 cm height) with open ends covered with cloth mesh and then, the tubes were hung at the geometrical center of the glass jars and sealed with air-tight lids. In the control jars, only acetone was applied on the filter papers. Jars were kept in the incubator and mortality was determined after 24, 48 and 72 h after exposure began. Insects were considered dead when no leg or antennal movements were observed and each experiment was replicated thrice for each concentration.

Antifeedant activity: The antifeedant bioassay of sub-lethal doses on 1-7 days-old adults was done as designated for fumigant toxicity with concentrations viz., LC_{25}-LC_{45}. Survival adults from 24 h fumigant bioassays were removed and used immediately for antifeedant assays. In each petri dish (6 cm) 10 g of wheat flour was poured and 50 treated alive insects with sub-lethal doses were transferred to it. The lids of petri dishes were pierced (1 cm diameter) and then covered by a fine mesh cloth for ventilation. Control groups were treated in the same way without oil. Each experiment was replicated four times in both control and treated groups. Decrease of flour weight in each petri dish was calculated after 48 h as follows:

$$\text{FDI (\%)} = \frac{\text{C-T}}{\text{C}} \times 100$$

where, FDI is the Feeding Deterrence Index, C is the consumption of insect without essential oil and T the consumption of insects treated with essential oil concentrations.

Statistical analysis: Mortality percentages were calculated by the Abbott's correction formula for natural mortality in the untreated control (Abbott, 1925). The experiments were arranged in a completely randomized design and the data were analyzed with ANOVA. The means were separated by the Turkey's test ($p = 0.05$). The lethal concentration (LC values), lethal time (LT values), 95% confidence intervals and chi-square were calculated using Probit analysis with statistical software SPSS version 16.0 (SPSS 16, IBM, Chicago, Illinois, US).

RESULTS AND DISCUSSION

Extraction and chemical composition of essential oil: Mean yield of extracted essential oil by Clevenger apparatus was 0.45%. Chemical analysis identified 14 compounds accounting for 99.13% of total essential oil. Decane (59.58%), dodecane (12.93%), ethylcyclohexane (6.84%), 5-methylnonane (4.71%), 3-methylnonane (4.32%) and tetradecane (3.82%) were main components in the *H. perforatum* essential oil (Table 1).

Recently the chemical compositions of essential oils of some *Hypericum* species from different countries have been studied. For example, in the study by Gudzic *et al.* (2001), (E)-anethole (30.7%) and β-farnesene (12.4%) in the *H. olympicum* L., essential oil and β-caryophyllene (14.2%) and 2-methyl-octane (13.1%) in the *H. perforatum* essential oil were found as main components. In the other study, a total of 74 components were identified by GC-MS in *H. scabrum* oil from Turkey, including α-pinene (9.26%), terpinen-4-ol (5.12%), camphor (5.94%), δ-cadinene (4.52%), pulegone (4.45%), γ-muurolene (4.12%), pinocarvone (3.97%) and β-caryophyllene (3.42%) as predominant components (Tozlu *et al*., 2011). Essential oil yield of *H. helianthemoides* (Spach) Boiss., *H. scabrum* and *H. perforatum* from Iran were 0.12, 0.20 and 0.21 mL/100 g dried material, respectively. The major constituents of the essential oils were α-pinene (12.52-49.96%), β-pinene (6.34-9.70%), (E)-β-ocimene (4.44-12.54%), β-caryophyllene (1.19-5.67%) and germacrene-D (2.34-6.92%) (Pirbalouti *et al*., 2014). In the present study, decane (59.58%), dodecane

Table 1: GC-MS analysis of essential oil of *Hypericum perforatum* from Iran

Compound	Retention time (min)	Percentage
n-hexanol	4.544	0.70
(Z)-3-methyl-4-nonene	5.998	0.60
5-methylnonane	6.734	4.71
3-methylnonane	7.060	4.32
Ethylcyclohexane	7.529	6.84
Decane	7.897	59.58
1,2,3-trimethylcyclohexane	8.123	1.05
sec-butylcyclohexane	8.799	0.63
2,6-dimethyldecane	12.450	0.68
1-hexyl-3-methylcyclopentane	13.459	1.61
Dodecane	13.726	12.93
L(-)-carvone	15.002	0.64
Tetradecane	19.145	3.82
Hexadecane	24.006	1.02
Total		99.13
Yield		0.45

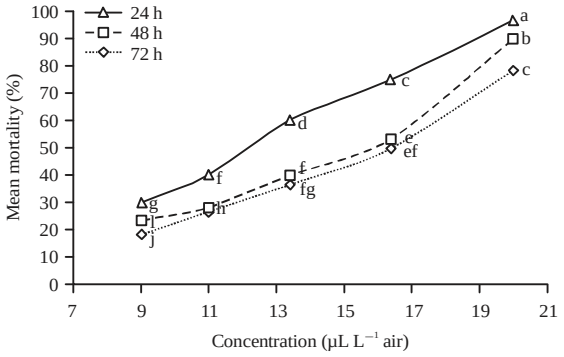

Fig. 1: Fumigant toxicity of the essential oil of *Hypericum perforatum* on *Tribolium castaneum* adults after 24, 48 and 72 h exposure times. Means with the same letters are not significantly different p = 0.05, Tukey's test

(12.93%), ethylcyclohexane (6.84%), 5-methylnonane (4.71%), 3-methylnonane (4.32%) and tetradecane (3.82%) were identified as main components of *H. perforatum* essential oil. Differences between mentioned results and results of present study can be attributed to plant species, season, location, climate, soil type, age of the leaves, soil fertility regimen, the drying method and the method of oil extraction (Batish *et al*., 2008; Rahimi-Nasrabadi *et al*., 2013).

Fumigant toxicity and antifeedant effect: Results of fumigant toxicity showed the adults of *T. castaneum* were very susceptible to the essential oil of *H. perforatum*. Toxicity depended on essential oil concentrations and exposure times ($F = 300.40$, $p<0.0001$ for essential oil concentration, $F = 69.28$, $p<0.0001$ for times and $F = 2.607$, $p = 0.027$ for concentrations-times) and highest levels of mortalities were achieved after 72 h of treatment. Comparison of the means by Tukey's test ($\alpha = 0.05$) also indicated that oil concentrations and exposure times have a statistically significant effect on the toxicity (Fig. 1).

Probit analysis indicated that lethal concentrations to kill 50% of insect population (LC_{50}) with their fiducial limits were 15.048 (13.949-16.496), 14.542 (13.405-15.990) and 11.743 (10.930-12.492) µL L^{-1} air in the 24, 48 and 72 h exposure times, respectively. Along with increasing of exposure time, the mortality rate increased and the LC_{50} values decreased from 15.048-11.743 µL L^{-1} air (Table 2). The LT_{50} value (the time needed to kill 50% of the population) was 10.110 h at the highest concentration (20 µL L^{-1} air) (Table 3).

Sub-lethal concentrations provided by fumigant toxicity within 24 h exposure time were used in the antifeedant bioassays against adults of *T. castaneum*; $LC_{25} = 10.741$, $LC_{30} = 11.578$, $LC_{35} = 12.412$, $LC_{40} = 13.258$ and $LC_{45} = 14.132$ µL L^{-1} air. Essential oil of *H. perforatum* had significant antifeedant effects on adults of *T. castaneum* and food consumption reduced based on essential oil extension ($F = 42.109$, $p<0.0001$) (Fig. 2). Feeding Deterrence Index (FDI) were 30.78, 48.69, 63.91, 82.61 and 95.78% for mentioned concentrations, respectively. This activity also depended on essential oil concentrations and the concentration of 14.132 µL L^{-1} air (LC_{45}) caused the highest feeding deterrence effect and lowest food consumption which were 95.78% and 0.097 g, respectively (Fig. 2).

Table 2: LC_{50} values and their related information in the fumigant toxicity test of *Hypericum perforatum* essential oil against *Tribolium castaneum*

Time (h)	LC_{50} with 95% confidence limits (µL L^{-1} air)	Intercept	Slope	χ^2 (df = 3)	p-value [a]
24	15.048 (13.949-16.496)	-5.424	4.607	3.360	0.339
48	14.542 (13.405-15.990)	-4.917	2.229	3.135	0.731
72	11.743 (10.930-12.492)	-6.87	5.877	4.570	0.206

[a]Since the significance level is greater than 0.150, no heterogeneity factor is used in the calculation of confidence limits, df: Degree of freedom

Table 3: LT_{50} values at high tested concentration for fumigant toxicity of *Hypericum perforatum* essential oil against *Tribolium castaneum*

Concentration (µL L^{-1} air)	LT_{50} with 95% confidence limits (h)	Intercept	Slope	χ^2 (df = 3)	p-values [a]
20	10.110 (1.004–17.513)	-2.048	2.029	0.297	0.586

[a]Since the significance level is greater than 0.150, no heterogeneity factor is used in the calculation of confidence limits, df: Degree of freedom

Some of *Hypericum* spp., from different countries have been evaluated for their insecticidal effects against insect pests. The Turkish *H. scabrum* L., essential oil was tested for toxicity against broad bean weevil (*Bruchus dentipes* Baudi). The essential oil was toxic to adults and insect mortality increased with increasing concentration of oil. Twenty microliters of essential oil caused 100% mortality after 36 h exposure time (Pirbalouti *et al*., 2014). The essential oils of *H. scabrum* and *H. perforatum* from Turkey were tested against adults of *Sitophilus granarius* (L.). Mortality rate of *S. granarius* adults increased significantly as the dosage level and exposure time increased. After 96 h of treatment, highest levels of mortalities (95.96%) were recorded at the dose of 1 µL essential oil of *H. perforatum* (Kordali *et al*., 2012). Larvicidal activity of essential oil from some Tunisian *Hypericum* spp., against *Culex pipiens* larvae were evaluated in the study of Rouis *et al*. (2013). One hundred and thirty four compounds were identified, ranging between 85.1 and 95.4% of the oil's composition. Results revealed essential oils of *Hypericum* spp., have a significant larvicidal activity against *C. pipiens* with LC_{50} ranging between 102.82 and 194.70 ppm. It is obvious that the essential oils isolated from *Hypericum* spp., especially *H. perforatum* have good insecticidal

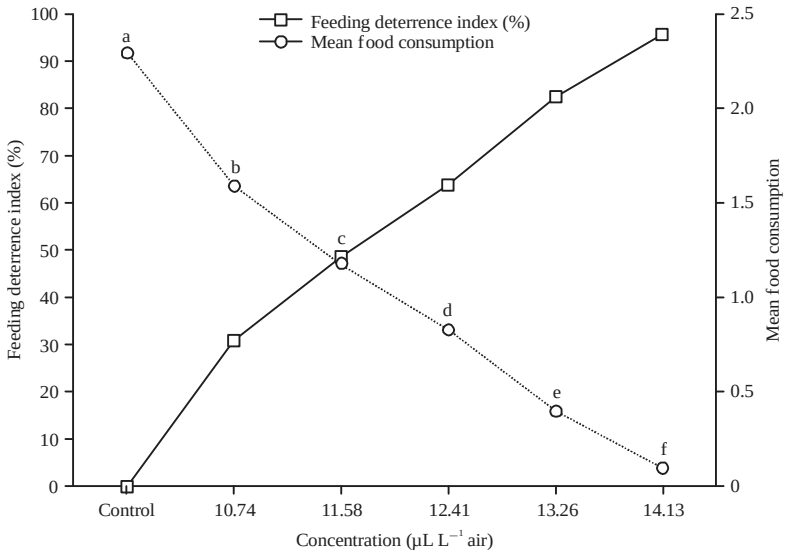

Fig. 2: Feeding deterrent effects of the essential oil of *Hypericum perforatum* against the treated adults of *T. castaneum* with sub-lethal concentrations from 24 h LC_{25}-LC_{45} (10.74-14.13 µL L^{-1} air). Mean food consumption with different letters are significantly different (p = 0.05, Tukey's test)

properties against insect pests and studies mentioned confirm results of the present work for toxicity of *H. perforatum* essential oil on insect pest.

Antifeedant activities of some plant essential oils have been evaluated in the literature. Huang *et al.* (2000) tested bioactivities of essential oil from cardamom, *Elletaria cardamomum* (L.) Maton., to *T. castaneum* and *Sitophilus zeamais* Motschulsky. Feeding deterrence studies showed that cardamom oil did not have any feeding deterrence effects on either adults or larvae of *T. castaneum*. However, it significantly reduced all the nutritional indices of the adults of *S. zeamais* but with very slight feeding deterrence (27%) at a concentration of 1.44×10^4 ppm. In another study, the essential oil extracted from *Carum copticum* C.B. Clarke (Apiaceae) was tested against *T. castaneum*, for antifeedant activity. In this study, several experiments were designed to measure the nutritional indices such as Relative Growth Rate (RGR), Relative Consumption Rate (RCR), efficiency of conversion of ingested food (ECI) and Feeding Deterrence Index (FDI). Results indicated that nutritional indices varied significantly as essential oil concentrations increased. *C. copticum* essential oil increased FDI as the oil concentration was increased (Sahaf and Moharamipour, 2009). In the present study, the antifeedant effect of essential oil of *H. perforatum* on *T. castaneum* was evaluated for the first time. Compared to conventional chemical methods, feeding inhibitors have several advantages in plant protection including the fact that the host choice of generalists and specialists may be modified when inhibitors are used. If an insect species can feed on other plants, it can be easier to direct away than if it is highly specialized in one host. Along with lethal treatment, the practice of using feeding inhibition allows us to develop and exploit naturally occurring plant defense mechanisms, thereby reducing the use of traditional pest management chemicals (Ebadollahi, 2013).

The toxicities of essential oil's main components and their relationship have been investigated by many researches (Toloza *et al.*, 2006; Suthisut *et al.*, 2011; Akhtar *et al.*, 2012). In general, essential oils are mixtures of various components and their effects may be either the result of a synergism of all components or could reflect only those of the main components (Regnault-Roger *et al.*, 2012; Rajendran and Sriranjini, 2008). Accordingly, the insecticidal activities of essential oil of *H. perforatum* can be attributed to its main components such as decane, dodecane, ethylcyclohexane and 5-methylnonane.

Results indicate that *H. perforatum* essential oil has a considerable potential for management of *T. castaneum*. However, for the practical application of essential oils the associated problems with these compounds such as rapid degradation and high cost must be resolved. To overcome the rapid degradation of essential oils for using as pesticides, new formulation and techniques such as micro-capsulation and controlled release technique should be established. Large quantity harvesting of plants may be the answer to the high cost problem. Further, field trials of plant essential oils should be done for their practical application.

SIGNIFICANCE STATEMENT

Biological effects of plant essential oils such as pesticidal, antioxidant and antiseptic activities have been investigated in the recent years. Essential oils have multiple mode of action on insect pests and insect pest's resistance to these materials has not reported yet. Further, the broad spectrum effects of essential oils including lethal and sub-lethal activities have been detected against different insect pests. It was found that the essential oils have very low toxicity on mammals and they have considerable potential in the human health aspects. In the present study, biological effects of *Hypericum perforatum* essential oil proved against a cosmopolitan stored-product insect pests; *Tribolium castaneum* Herbst.

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