Insecticidal Effects of Peppermint and Black Pepper Essential Oils against Rice Weevil, *Sitophilus oryzae* L. and Rice Moth, *Corcyra cephalonica* (St.)

Khani M (Ph.D.)[1*], Muhamad Awang R (Ph.D.)[2], Omar D (Ph.D.)[2]

1- Cultivation & Development Department of Medicinal Plants Research Center, Institute of Medicinal Plants, ACECR, Karaj, Iran
2- Department of Plant Protection, Faculty of Agriculture, University Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
* Corresponding author: Cultivation & Development Department of Medicinal Plants Research Center, Institute of Medicinal Plants, ACECR, P.O.Box: 31375-1369, Karaj, Iran
Tel: +98-26- 34764010-18, Fax: +98-26- 34764021
Email: khanimousa@yahoo.com

Received: 13 Feb. 2012 **Accepted:** 24 Oct. 2012

Abstract

Background: **The rice weevil, *Sitophilus oryzae* and rice moth, *Corcyra cephalonica* are major and cosmopolitan insect. Adults of *S. oryzae* (male and female) and both larvae are insatiable feeders on a great variety of grains.**

Objective: **This study was conducted to estimate the insecticidal effect of essential oils from peppermint, *Mentha piperita* L. and black pepper, *Piper nigrum* L. against two major stored product insects.**

Methods: **Essential oils from two species of plants were obtained by clevenger-type water distillation. The major compounds in these essential oils were identified using gas chromatography-mass spectrometry and their insecticidal effect was tested against adults of the rice weevil, *Sitophilus oryzae* L. and the 3rd instars larvae of rice moth, *Corcyra cephalonica* (St.).**

Results: **The major compounds found in peppermint were menthol, isomenthone, limonene and cineole and in black pepper were limonene, α and β pinene and caryophyllene. Highest toxicities were observed against *S. oryzae* populations treated with *M. piperita* and *P. nigrum* essential oils with LC_{50} values of 85.0 and 287.7 µL/L air after 72 hours after commencement, respectively. In the case of *C. cephalonica* larvae, the LC_{50} values were 343.9 and 530.5 µL/L air for *M. piperita* and *P. nigrum* essential oils at 72 hours after commencement, respectively. These results are attributed to the compounds present in essential oils of *M. piperita* and *P. nigrum*.**

Conclusion: **It was resulted that *M. piperita* and *P. nigrum* oils have insecticidal effects against *S. oryzae* and *C. cephalonica*. For this reasons, the selected plant oils have potential for development of novel insecticides.**

Keywords: ***Sitophilus oryzae*, *Corcyra cephalonica*, Essential oil, *Mentha piperita*, *Piper nigrum*, Bioassay**

Introduction

The rice weevil, *Sitophilus oryzae* is a major and cosmopolitan insect. Both adults (male and female) and larvae are insatiable feeders on a great variety of grains. They attack wheat, corn, rice, oats, rye, barley, dried beans, sorghum and cereal products, especially macaroni and females deposit the eggs within seed grains [1, 2]. The damage by *S. oryzae* to corn crops in South America was estimated 10%, with an approximate value of 30 billion dollars each year [3, 4]. The rice moth, *Corcyra cephalonica* (St.) is another widespread pest of stored food commodities. Their larvae cause damage by feeding on the stored products. They feed on rice, maize and other cereals in storage [5, 6]. In addition, the larvae also cause extensive indirect quantitative and qualitative damage by making durable silk webs, fecal material and leaving threads of silk when they shift and the stored grains are contaminated by frass, excreta and pupal cocoons [4, 7]. Use of synthetic fumigant materials are the most widespread selected method in the control of stored product pests. Due to the ease and ability to control most stored product pests, methyl bromide (MeBr) and phosphine (PH3), are widely used as fumigants. Due to ozone depleting properties in the atmosphere, the use of methyl bromide has been restricted in most developed countries since 2005, and is used in emergency situations and for quarantine purposes only [8, 9]. In accordance with the Montreal Protocol the Chinese government has also completely banned the use of methyl bromide in cereal production since end of 2006 [9]. Now, the control of stored insect pests is heavily dependent on the use of phosphine. But, using of phosphine as a fumigant over long periods have resulted in many problems such as insect pest resistance, toxicity to mammals, residue problems, other non-target organisms, and decrease in natural enemies [10, 11]. Insect resistance to phosphine has been reported from more than 45 countries [12].

A large amount of powders, extracts and essential oils from natural products, believed to be non hazardous to humans and the environment, have been employed as biopesticides to control different insect pests. It must be noted that plant materials have been used as insecticides against different economically important pests before the invention of chemical pesticides such as DDT [13]. There is a revival in the use of botanical insecticides for insect pest control to minimize environmental pollution, and hence there is the need to find potential natural products for storage product pest control. Botanical insecticides are unique in action and can be widely used in pest control and many are safe for use on stored food commodities to protect against damage or losses due to insect infestation [5, 14]. Over the past 15 years, interest in botanical insecticides has increased as a result of environmental concerns and insect populations becoming resistant to conventional chemicals [15]. Botanical insecticides are naturally occurring insecticides that are derived from plants [16]. Essential oils from different plant species possess ovicidal, larvicidal and repellent effects against various insect species and are regarded as environmentally compatible pesticides [16, 17]. Due to limited research has been conducted on some biological activities such as toxicity, antifeedant, repellency and inhibition of adult emergence effects of essential oils from peppermint, *Mentha piperita* and black pepper, *Piper nigrum* against rice weevil, *Sitophilus oryzae,* also there is very little documentation on the rice moth, *Corcyra cephalonica*, this study was conducted to determine the chemical constituents of essential oils from *Mentha piperita* and *Piper nigrum* and evaluate insecticidal effect of essential oils against the adults of *Sitophilus oryzae* and 3rd instar larvae of *Corcyra cephalonica*.

Material and Methods

Colonies of rice weevil, *S. oryzae* and rice moth, *C. cephalonica* were obtained from the Entomology laboratory stock culture of University Putra Malaysia (UPM). Rice weevil were reared on sterilized whole rice grains that stored at -15 °C for two weeks to annihilate any previous insects [18] with 13-14% initial moisture content in laboratory conditions [19, 20] in containers. Rice moth, *C. cephalonica* were reared on medium consisting of finely ground rice and maize flour in the ratio 1:1 (w/w) under laboratory conditions (27 ±1 ˚C, 75 ± 5% R.H. with a 12:12 h light: dark cycle). The food media were sterilized in an autoclave before experimentation. The subcultures and the tests were carried out under the same conditions, and experiments were conducted on 7-14 day old adults of *S. oryzae* due to obtain of the same age of insects and third instars larvae of *C. cephalonica*. All containers were placed on trays with water to prevent entry of any crawling insects [7, 21].

Plant materials

The fresh fruits of *P. nigrum* and fresh herbage of *M. piperita* were washed in tap water and placed separately into 2 L round bottom flasks. Distilled water was added into round bottom flasks until all the plant materials were completely immersed. The flasks were then heated in a clevenger-type apparatus heater and the mixture was boiled for 3 hours to evaporate the components in the plant materials. Anhydrous sodium sulphate was used to remove traces of water. The essential oils obtained were stored in a refrigerator at 4 ˚C [22, 23] to avoid of degradation of components until used.

The GC and GC-MS analysis was carried out in Institute of Medicinal Plants, ACECR, to determine the major components of the volatile oils. The essential oils from *M. piperita* and *P. nigrum* were separated by gas chromatography using a 30 m × 0.25 mm (0.25 µm film thickness) HP-5 capillary column. One micro litter samples were injected into the chromatograph using an auto sampler, equipped with a split/splitless injector and a flame ionization detector (FID). The injector temperature was programmed at 290 ^{o}C. The column temperature programme initiated runs at 50 ^{o}C, for 5 min, warmed to 240 ^{o}C at 15 ^{o}C /min, warmed to 300 ^{o}C then held for 3 min to facilitate optimal separation and helium as carrier gas at a flow rate of 0.8 ml min^{-1}.

Also to evaluate oils constituents the prepared oils were analyzed using a GC-MS system (Agilent 5973) equipped with a HP-5MS column (30 m × 0.25 mm × 0.25 µm), and helium as carrier gas at a flow rate of 1 ml min^{-1}, in electronic impact mode (70 eV) and split injection ratio (1:20). The injector and GC-MS interface were kept at 300 °C. The column temperature programme was as above. The components of the oils were identified by comparison of the mass spectra stored in the NIST08 computer database library information and/or the mass spectra published in authentic literature.

Toxicity bioassay

Toxicity tests of the plant essential oils against *S. oryzae* adults and *C. cephalonica* larvae were carried out in the laboratory according to the methods described by Allotey and Azalekor [7] and Negahban *et al* [12]. To evaluate the fumigant toxicity effects of essential oils from *M. piperita* and *P. nigrum* against adults of *S. oryzae*, essential oils at 2, 4, 6, 8 and 10 µL quantities were dissolved in 1 mL acetone to give dose ranges of 74 to 370 µL/L air (74, 148, 222, 296 and 370 µL/L air). The prepared treatments were applied on filter papers (Whatman No. 1, cut into 2 cm diameter pieces). After evaporating the solvent the impregnated filter papers were attached inside screw caps of 27 ml glass vials [23]. Rice was added into the vials and 10 *S. oryzae* (7-14 days old) adults were released into the vials. The caps were tightly screwed on and the vials were sealed with parafilm. To determine the fumigant toxicity effects of essential oils from *M. piperita* and *P. nigrum* against 3rd instar larvae of *C. cephalonica*, amounts of 5, 10, 15, 20 and 25 µL of essential oils were dissolved in 1 mL acetone to give dose ranges of 200 to 1000 µL/L air (200, 400, 600, 800 and 1000 µL/L air). The treatments were applied on to filter papers (Whatman No. 1, 5 cm dia.), and impregnated filter papers were attached inside the Petri-dish (25 ml) covers and some rice was placed in the Petri-dishes. Twenty 3rd instar larvae were released in each Petri-dish and sealed with parafilm. The all experiments were conducted in five replicates for each treatment. The mortality was recorded after 24, 48 and 72 hours to determine LC_{50} values. In fumigant experiments, new groups of both insects were prepared each time and mortality for each exposure time was determined independently.

Results

Chemical composition

The essential oil from *P. nigrum* exhibited large amounts of monoterpenes derivatives and very low level of sesquiterpenes and oxygenated sesquiterpenes. The monoterpene component was made up of more than 93.5% of total essential oils, followed by sesquiterpenes (4.3%) and oxygenated sesquiterpenes (0.4%), while other components were at very low quantities.

Limonene, α-pinene and β-pinene appeared as the main components with 33.8%, and 31.2% and 23.3% of total essential oil from fresh fruits of *P. nigrum*, respectively (Table 1). These results indicate much higher values compared to Singh *et al.*, [24], who observed the presence of 49 compounds which made up 99.4% of total oil, with the following components, α-pinene (4.8%), β-pinene (6.7%), myrcene (0.9%), limonene (16.9%), terpinolene (0.2%), linalool (0.3%), terpinen-4-ol (2.0%) and β-caryophyllene (24.2%). Jirovetz *et al.*, [25] investigated compounds in essential oils of dried fruits of black pepper, *P. nigrum* from the Cameroon, and observed the presence of germacrene D (11.01%), limonene (10.26%), β-pinene (10.02%), α-phellandrene (8.35%), β-caryophyllene (7.29%), α-pinene (6.40%) and cis-β-ocimene (3.19%). These results were also different compared with the present study. In the present study, the essential oils of fresh fruits of *P. nigrum* had significantly larger amounts of limonene (33.8%), α-pinene (31.2%) and β-pinene (23.3%) while Pino *et al.*, [26], Singh *et al.*, [24] and Jirovetz *et al.*, [25] reported much lower concentrations of limonene (19.0%, 16.88% and 10.26%), α-pinene (8.2%, 4.74% and 6.40%) and β-pinene (12.0%, 6.71% and 10.02%), respectively. However, Singh *et al.*, [24] and Jirovetz *et al.*, [25] reported higher concentrations of caryophyllene (24.24% and 7.29%) than in the present study (4.06%). These variations could be attributed to climate, soil composition, plant organ, age and vegetative conditions [27].

The essential oil of *M. piperita* extracted by hydrodistillation included monoterpenes (10.7%), high percentage of oxygenated monoterpenes (87.6%), followed by a small amount of sesquiterpenes (1.3%). Thirty-eight components were identified from GC-MS analysis in essential oil of peppermint, *M. piperita*, and the major known components are presented in Table 2. The major oxygenated monoterpene compounds were menthol (46.9%), menthone (19.8%), cineol (5.40%), cyclohexanone (5.4%) and isomenthol acetate (2.8%), and limonene (7.5%) appeared as the main monoterpene compound (Table 2). Similar results were reported by Regnault-Roger *et al.* [28].

Table 1- Major constituents of *P. nigrum* fresh fruit essential oil (GC-MS Analysis)

Component	Formula	Mol. Weight	Retention time (min)	% in total oil (v/v)
α-Pinene	$C_{10}H_{16}$	136	10.212	31.18
β-Pinene	$C_{10}H_{16}$	136	9.380	23.34
β-Myrcene	$C_{10}H_{16}$	136	9.622	3.91
Limonene	$C_{10}H_{16}$	136	10.858	33.84
Caryophyllene	$C_{15}H_{24}$	204	20.698	4.06

Table 2- Major constituents of *M. piperita* essential oil (GC-MS Analysis)

Component	Formula	Mol. Weight	Retention time (min)	% in total oil (v/v)
Limonene	$C_{10}H_{16}$	136	10.746	7.49
Cineole	$C_{10}H_{18}O$	154	10.853	5.40
Isomenthone	$C_{10}H_{18}O$	154	14.431	19.85
Cyclohexanone, 5-methyl-2-(-,ethylethyl)-	$C_{10}H_{18}O$	154	14.638	5.37
Menthol	$C_{10}H_{20}O$	156	15.091	46.98
Isomenthol acetate	$C_{12}H_{22}O_2$	198	17.553	2.78

Toxicity effects

Results on toxicity effects of the essential oils showed significant differences between the different concentrations against adults of *S. oryzae* and 3rd instar larvae of *C. cephalonica* (Figure 1). Highest toxicities were observed against *S. oryzae* populations treated with *M. piperita* and *P. nigrum* essential oils with LC_{50} values of 85.0 and 287.7 µL/L air after 72 hours after commencement, respectively (Table 3). In the case of *C. cephalonica* larvae, the LC_{50} values were 343.9 and 530.5 µL/L air for *M. piperita* and *P. nigrum* essential oils at 72 hours after commencement, respectively. These results are attributed to the compounds present in essential oils of *M. piperita* and *P. nigrum*. The results showed that *M. piperita* and *P. nigrum* essential oils were relatively more toxic to adults of *S. oryzae* than 3rd instar larvae of *C. cephalonica*. At the 370 µL/L air dose level, *M. piperita* and *P. nigrum* essential oils caused 97% mortality of *S. oryzae* adults within 72 hours of exposure (Figure 1), while *M. piperita* and *P. nigrum* essential oils caused 40.2 and 59.6% mortality to 3rd instar larvae of *C. cephalonica* for the same duration of exposure with 400 µL/L air, respectively. Mortalities of *C. cephalonica* larvae reached to 91 and 76.3% at the highest concentration (1000 µL/L air) with *M. piperita* and *P. nigrum* essential oils, within 72 hours exposure, respectively (Figure 2).

Discusion

Insecticidal, antimicrobial and antifungal activities of limonene, α- and β-pinene have been observed [29-33]. Insecticidal activity of limonene has been reported against the early 4th instar larvae of the mosquito *Culex auinquefasciatus*. The LC_{50} values after 24 and 48 h were determined as 53.80 and 32.52 ppm, respectively [30]. Hebeish *et al.* [31] noted that limonene is used in food commodities due to its lemon-like flavour and odour, and also in soaps and perfumes. Limonene is also an active ingredient in 15 pesticide products used as insecticides, insect repellents, and dog and cat repellents. Jang *et al.*, [32] and Phillips *et al.*, [34] reported that limonene, α-pinene and

Table 3- Fumigant toxicity of *P. nigrum* and *M. piperita* essential oils against adult *Sitophilus oryzae* and 3rd instar larvae of *Corcyra cephalonica* at 72 hours after commencement of exposure

Insect Species	Treatment	Slope ± SE	Chi square (x^2)	Df[1]	LC_{50} (µL/L air) (Min-Max)[2]
S. oryzae	*M. piperita*	2.75±0.42	3.40	3	85.04 (59.59±106.37)
	P. nigrum	4.29±0.53	2.36	3	287.70 (239.65-331.08)
C. cephalonica	*M. piperita*	2.93±0.39	1.85	3	343.96 (276.92-402.63)
	P. nigrum	2.47±0.40	1.57	3	530.53 (444.58-625.50)

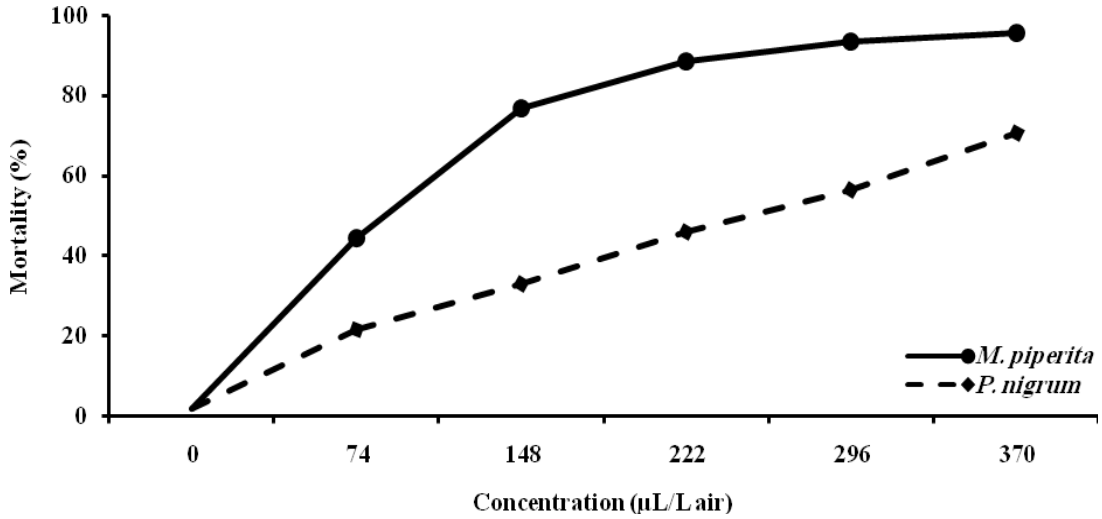

Figure 1- Mean mortality of *Sitophilus oryzae* adults at 72 hours after exposure to essential oils

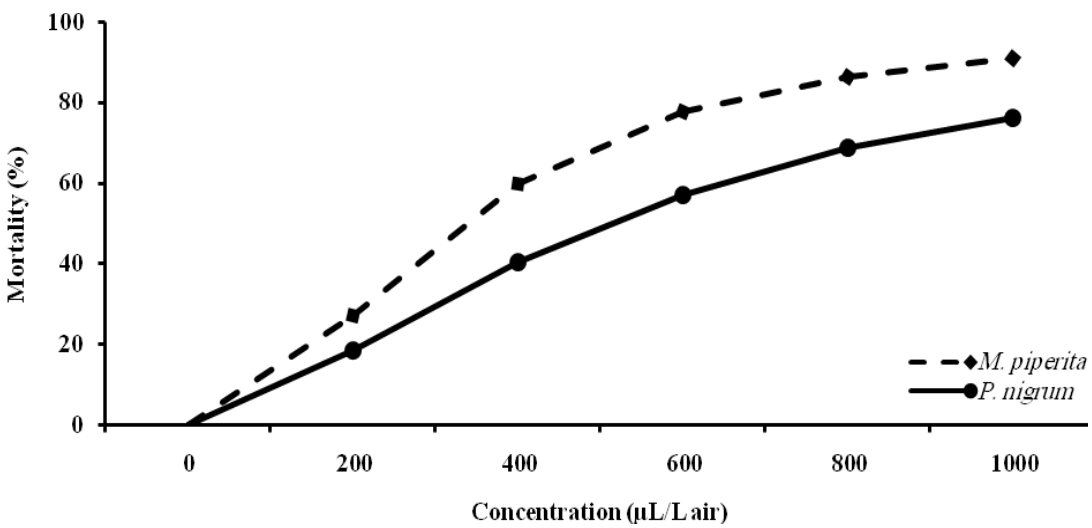

Figure 2- Mean mortality of *Corcyra cephalonica* adults at 72 hours after exposure to essential oils

β-pinene are used as a fumigant with contact toxicity against *Blattella germanica*. The LC_{50} for limonene was 2.58 mg/cm^2 and LD_{50} for α-pinene and β-pinene were 218.17 and 143.76 mg/L, respectively. Jung *et al.*, [35] found that IR-(+)-α-pinene (LC_{50}=0.36 mg/cm^2) and IR-(+)-β-pinene (LC_{50}=0.12 mg/cm^2) were the most toxic against the female German cockroach, *Blattella germanica* and can cause 100% mortality. Lee *et al.*, [33] reported that limonene can cause 100% mortality in housefly, German cockroach, rice weevil and saw-toothed grain beetle. The insecticidal activities of many essential oils and some monoterpenes have been also demonstrated against different insect species [29, 33, 36 - 40]. LC_{50} values of menthone, linalool and α-pinene against *S. oryzae* were 12.7, 39.2 and 54.9 µl/L air, respectively [41]. The toxicity of menthone against adult Colorado potato beetles was reported by Kordali *et al.* [38]. Who has been reported 100% mortality of Colorado potato beetle with limonene, α-pinene and β-pinene.

The plants of Lamiaceae family have been widely acknowledged to have pesticidal constituents. The essential oil from *Mentha* species including secondary metabolite components such as menthol, menthone and limonene have been studied for their insecticidal activity against several stored product pests [47, 58, 61 - 63]. The essential oils extracted from *M. piperita* have also been reported as a source of botanical insecticides [64]. The present research showed that the major components of essential oil from *M. piperita* were menthol (46.98%), isomenthone (19.85%) and limonene (7.49%). The essential oils of *M. piperita* showed strong insecticidal activity (LC_{50}=7.5 µl/L air) against *S. oryzae* [63]. The lower toxicity effect observed in the present study could be attributed to the different strain of *S. oryzae* or differences in composition of *M. piperita* essential oil employed in the two studies [61]. This observation is in agreement with that of Michaelraj *et al.* [58] who reported complete mortality of *S. oryzae* at 100 and 150 µl/250 ml of *M. piperita* oil, but *C. cephalonica* was not sensitive to *M. piperita* extract at 5µl/250 ml after 48 hours of exposure.

The Piperaceae family has been reported to have insecticidal activities due to presence of many potential phytochemicals. The *P. nigrum* extracts offer a unique and beneficial source of bio-pesticide material for the control of insect pests on a small scale [42, 43]. Many insecticidal components of plant extracts are mainly monoterpenes such as limonene which have been shown to be toxic to *Tribolium castaneum* [44, 45]. The toxic effect of *P. nigrum* was reported against some test insects. *P. nigrum* was shown to be most toxic to *Callosobruchus chinensis*, *Acanthoscelides obtectus*, *C. cephalonica*, *Ephestia cautella* Hubn., followed by *Oryzaephilus surinamensis* (L.), *Sitophilus zeamais* Mosteh, *Rhyzopertha dominica* (Fab.) and *Tribolium castaneum* Herbst [46]. The high toxicity effects of *P. nigrum* essential oils against *S. oryzae* adults and 3rd instar larvae of *C. cephalonica* are attributed to the presence of high concentrations of well-known toxic components such as caryophyllene and piperine.

The insecticidal components of a great number of plant extracts and essential oils are mostly monoterpenoids and sesquiterpenes [28]. Monoterpenoids have strong toxicity to insects due to high volatility, and lipophilic properties can penetrate into insects rapidly and interfere in physiological functions [12, 47]. Limonene is a monoterpenoid with various toxic activities [30, 48-51] including neurotoxic effects [52] and inhibition of reproduction and growth regulatory effects in several species of insects [53, 54]. The toxicity of plant essential oils are attributed to the major active component present. The fumigant toxicity effects of plant essential oils have also been widely reported [9, 22, 47, 55-60]. However, in the present study, the adults of *S. oryzae* and 3rd instar larvae of *C. cephalonica* were not very sensitive to *P. nigrum* essential oils compared with *M. piperita* essential oil (Table 3).

Conclusion

Based on results, *M. piperita* and *P. nigrum* oils showed toxic effects on two serious insect pests of stored grains rice weevil, *S. oryzae* and rice moth, *C. cephalonica*. Due to easily available of these products, the farmers and general public can use these oils in order to protect of stored grains, which are non-toxic in handling and use. For this reasons, the studied oils have potential for development of Bio-insecticides.

Acknowledgements

The authors thank to Institute of Medicinal Plant, ACECR, Tehran, Iran for the assistance to supply the plant essential oils and their analysis.

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