Tropical Journal of Pharmaceutical Research February 2013; 12 (1): 99-103
ISSN: 1596-5996 (print); 1596-9827 (electronic)
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Available online at http://www.tjpr.org
http://dx.doi.org/10.4314/tjpr.v12i1.16

Original Research Article

Chemical Composition and Insecticidal Activity of the Essential Oil of the Aerial Parts of *Ostericum grosseserratum* (Maxim) Kitag (Umbelliferae)

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Received: 26 March 2012 *Revised accepted:* 24 November 2012

Abstract

Purpose: *To investigate the chemical composition and insecticidal activity of the essential oil of the aerial parts of Ostericum grosseserratum against the maize weevil, Sitophilus zeamais.*
Methods: *Steam distillation of the aerial parts of O. grosseserratum during the flowering stage was carried out using a Clavenger apparatus in order to obtain its volatile oil content. Gas chromatography/mass spectrometric (GC/MS) analyses (HP-5MS column) of the essential oil were performed and its contact and fumigant activity determined.*
Results: *A total of 43 components of the essential oil were identified. The principal compounds were (d)-limonene (16.2 %), 4-terpineol (13.5 %), myristicin (11.3 %), γ-terpineol (8.3 %), β-pinene (5.1 %), β-caryophyllene (4.6 %) and linalool (4.1 %). The oil exhibited contact toxicity against adult S. zeamais with lethal concentration (LC_{50}) value of 17.97 µg/adult. The essential oils also possessed fumigant toxicity against S. zeamais with LC_{50} value of 13.70 mg/L air.*
Conclusion: *The study indicates that the essential oil of O. grosseserratum has a potential to be developed into a natural fumigant/insecticide for the control of grain storage insects.*

Keywords: *Ostericum grosseserratum; Sitophilus zeamais; Insecticidal, Contact toxicity; Fumigant; Essential oil*

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INTRODUCTION

The genus *Ostericum* belongs to the family Umbelliferae and is distinguished from the Genus *Angelica* by the presence of high concentrations of flavonoids in the leaf and mericarp of *Ostericum* [1]. It comprises only of about 10 species in the world, of which seven are distributed in China [2]. *Ostericum grosseserratum* (Maximowicz) Kitagawa is a herbaceous plant distributed mainly in some areas of China, Korea, Japan, Russia as well as Mongolia [2,3]. The roots of *O. grosseserratum* have reputed medicinal value as a regional substitute for the traditional Chinese medicine, "Radix Angelicae Biseratae" (*Angelica biserrata* or *A. pubescens*) [2,3]. This medicinal herb was used in traditional Chinese medicine as an analgesic and anti-inflammatory agent in the treatment of rheumatism and rheumatoid arthritis [4].

In a previous study, 8 constituent compounds (myristic acid, palmitic acid, stearic acid,

octacosanoic acid, β-sitosterol, isoscopoletin, succinic acid, and β-sitosterol-β-D-glucoside) were isolated from the ethanol extract of *O. grosseserratum* roots [3]. The chemical composition of the essential oils derived from *O. grosseserratum* roots has also been determined [5]. During the screening program for new agrochemicals from Chinese medicinal herbs and wild plants, the essential oil of *O. grosseserratum* flowering aerial parts was found to possess insecticidal toxicity against the maize weevil, *Sitophilus zeamais* Motsch.

However, a literature survey shows that there is no report on the chemical composition and insecticidal activity of the essential oil derived from the aerial parts of *O. grosseserratum*. Thus, the objective of this study was to investigate the chemical constituents and insecticidal activity of the essential oil of *O. grosseserratum* aerial parts against grain storage insect.

EXPERIMENTAL

Plant collection and identification

The aerial parts of *O. grosseserratum* were collected in August 2009 during the flowering stage from Xiaolongmen National Forest Park (39.48° N latitude and 115.25° E longitude, Mentougou District, Beijing 102300). The samples were air-dried and identified by Dr. Liu, (College of Life Sciences, Beijing Normal University, Beijing 100875, China) and a voucher specimen (no. ENTCAU-Umbelliferae-10003) was deposited at the Department of Entomology, China Agricultural University (Beijing 100193).

Extraction and isolation of essential oils

The flowering aerial parts were air-dried and first ground to powder using a grinding mill (Retsch Muhle, Germany). The powder was hydro-distilled for 6 h in a Clavenger apparatus. The oil was dried over anhydrous Na_2SO_4 and kept in a refrigerator (4 °C) pending subsequent experiments.

Analysis of the essential oils

Capillary gas chromatography was performed using Hewlett–Packard 5890 gas chromatograph equipped with a flame ionization detector and fused silica capillary column HP-5 (5 % diphenyl and 95 % dimethylpolysyloxane, 30 m × 0.25 mm, 0.25 µm film thickness), at a flow rate of 1 mL min^{-1}. Temperature was programmed from 60 to 280 °C (at a rate of 2 °C min^{-1}); injector and detector temperatures were 270 ^{o}C and 300 °C, respectively**.** The components of the essential oils were separated and identified by gas chromatography–mass spectrometry (GC–MS) using Agilent 6890N gas chromatography coupled to Agilent 5973N mass selective detector. The system was equipped with a flame ionization detector and capillary column with HP-5MS (30m × 0.25mm × 0.25µm). GC settings were as follows: the initial oven temperature was held at 60 °C for 1 min and ramped at 10 °C min^{-1} to 180 °C where it was held for 1 min, and then ramped at 20 °C min^{-1} to 280 °C and held there for 15 min. The injector temperature was maintained at 270 °C. The samples (1 µL) were injected neat, with a split ratio of 1:10. The carrier gas was helium at a flow rate of 1.0 ml min^{-1}. Spectra were obtained over the scan range 20 to 550 m/z at 2 scans s^{-1}. Most constituents were identified by gas chromatography by comparison of their retention indices with those published in the literature or with those of authentic compounds available in our laboratories. The retention indices were determined in relation to a homologous series of *n*-alkanes (C_8–C_{24}) under the same operating conditions. Further identification was made by comparison of their mass spectra with those stored in NIST 05 and Wiley 275 libraries or with mass spectra from literature [6]. Relative percentages of the oil components were calculated based on GC peak areas without using correction factors.

Insects

S. zeamais was obtained from laboratory cultures maintained for the last 10 years in the dark in incubators at 27-29 °C and 70 – 80 % relative humidity. Adult *S. zeamais* insects were reared on whole wheat at 12 – 13 % moisture content in glass jars (diameter 85 mm, height 130 mm). Unsexed adults of the insects used in all the experiments were about one week old. All containers housing insects and the petri dishes used in experiments were made escape-proof with a coating of polytetrafluoroethylene (Fluon, Blades Biological, UK).

Fumigant toxicity test

A Whatman filter paper (diameter 2.0 cm, CAT no. 1001020) was placed on the underside of the screw cap of a glass vial (diameter 2.5 cm, height 5.5 cm, volume 24 mL). Range-finding studies were run to determine the appropriate testing concentrations. Ten microliters of essential oil solution was added to the filter paper. The solvent was allowed to evaporate for 15 s before the cap was placed tightly on the glass vial (containing 10 unsexed insects) to form a sealed chamber. Fluon (ICI America Inc) was used inside the glass vial to prevent insects from

the treated filter paper. *n*-Hexane was used as controls and six replicates were used in all treatments and controls. They were incubated at 27 – 29 °C and 70 – 80 % relative humidity for 24 h and mortality of insects was observed.

Contact toxicity test using topical application

The contact toxicity of the essential oil against adult *S. zeamais* was measured as described by Liu and Ho [10]. Range-finding studies were run to determine the appropriate testing concentrations. A serial dilution of the essential oil was prepared in *n*-hexane. Aliquots of 0.5 µl of the dilutions were applied topically to the dorsal thorax of the insects. Controls were determined using *n*-hexane. Six replicates were used in all treatments and controls. Both treated and control insects were then transferred to glass vials (10 insects/vial) with culture media and kept in incubators. Mortality of insects was observed after 24 h. Pyrethrum extract (25% pyrethrine I and pyrethrine II) was purchased from Fluka Chemie.

Statistical analysis

The results from all replicates in fumigant and contact toxicity were subjected to Probit analysis [7] using PriProbit Program V1.6.3 to determine LD_{50} and LC_{50} values, respectively [8]. Samples for which the 95 % fiducial limits did not overlap were considered to be significantly different.

RESULTS

The yellow essential oil was yellow with a yield of 0.12 % (v/w) and density of 0.83 g/ml. A total of 43 components of the essential oil were identified, accounting for 94.7 % of the total oil. The principal compounds in the essential oil were (d)-limonene (16.2 %), 4-terpineol (13.5 %), myristicin (11.3 %), γ-terpineol (8.3 %), and β-pinene (5.1 %), followed by β-caryophyllene (4.6 %) and linalool (4.1 %) (Table 1). Monoterpenoids represented 22 of the 43 compounds, corresponding to 60.5 % of the whole oil while 29 of the 43 constituents were sesquiterpenoids (21.3 % of the whole essential oil). The essential oil exhibited contact toxicity against *S. zeamais* adults with an LC_{50} value of 17.97 µg/adult (Table 2). The oil also showed fumigant toxicity against maize weevils with an LC_{50} value of 13.70 mg/L (Table 2).

DISCUSSION

The main constituents of the essential oil of *O. grosseserratum* flowers were (d)-limonene, 4-terpineol, myristicin, γ-terpineol and β-pinene followed by β-caryophyllene and linalool. Its chemical composition is different from that derived from the roots of the same plant in a previous report [5] contains octanal (5.9 %), β-pinene (5.6 %), neoisothujyl alcohol (4.6 %), γ-patchoulene (3.9 %), ρ-cymene (3.5 %), α-pinene (3.4 %), heptanal (3.2 %) and 3,7-dimethyl-1-octene (3.0 %) as major constituents. This suggests that there is a great variation in chemical composition of the essential oils derived from different parts of *O. grosseserratum*.

The essential oils of *O. grosseserratum* exhibited contact toxicity against *S. zeamais* adults. However, compared with the positive control (*Pyrethrum* extract, 25 % pyrethrine I and pyrethrine II) with an LC_{50} value of 4.3 µg/insect [9], acute toxicity against the weevil was weak. The essential oil also showed fumigant toxicity against adult *S. zeamais*. The commercial grain fumigant, methyl bromide (MeBr), is reported to possess fumigant activity against *S. zeamais* adults with an LC_{50} value of 0.67 mg/L [10]. Thus, the essential oil is 20 times less toxic to adult *S. zeamais* than with MeBr.

However, compared with the fumigant activity of the other essential oils reported in the literature and which were tested using a similar bioassay, the essential oil obtained in the present study exhibited the same or stronger fumigant toxicity against maize weevils, e.g., the essential oils of *Artemisia lavandulaefolia* (LC_{50} = 11.2 mg/L) and *A. sieversiana* (LC_{50} = 15.0 mg/L) [11], *Illicium fragesii* and *I. simonsii* (LC_{50} = 11.36 mg/L and 14.95 mg/L, respectively) [12,13], and *Kadsura heteroclita* (LC_{50} = 14.04 mg/L) [14] and *Ostericum sieboldii* (LC_{50} = 20.92 mg/L) [15].

The foregoing suggest that the fumigant activity of the essential oil of *O. grosseserratum* has some promise as a possible natural fumigant/insecticide for the control of grain storage insects ,especially as currently used fumigants are synthetic insecticides and the most effective fumigants are also highly toxic to humans and other non-target organisms [16]. However, to develop a practical application for the essential oil as novel fumigant/insecticide, further research into the safety of the essential oil in humans is needed. Additional studies on the development of formulations are also necessary to improve efficacy and stability as well as to reduce cost.

CONCLUSION

The essential oil of the flowering aerial parts of *O. grosseserratum* demonstrated some activity

Table 1: The main compounds of the essential oil of *Ostericum grosseserratum*

Peak no.	Compound	Retention index	(%)
1	α-Pinene	931	1.2
2	Camphene	952	0.3
3	**β-Pinene**	**974**	**5.1**
4	β-Myrcene	993	0.2
5	α-Terpinene	1017	0.4
6	ρ-Cymene	1024	1.5
7	β-Phellandrene	1026	2.1
8	**d-Limonene**	**1027**	**16.2**
9	1,8-Cineol	1031	1.2
10	**γ-Terpinene**	**1057**	**8.3**
11	**Linalool**	**1097**	**4.1**
12	Thujanol	1146	0.4
13	Borneol	1167	1.3
14	**4-Terpineol**	**1177**	**13.5**
15	p-Cymene-8-ol	1182	0.1
16	α-Terpineol	1189	0.4
17	Verbenone	1204	1.0
18	*cis*-Geraniol	1229	0.7
19	Eucarvone	1245	0.3
20	*trans*-Geraniol	1252	1.3
21	Bornyl acetate	1285	0.5
22	Cubebene	1352	0.1
23	Eugenol	1356	1.6
24	Ylangene	1372	0.3
25	Copaene	1375	1.7
26	β-Elemene	1390	0.8
27	**β-Caryophyllene**	**1420**	**4.6**
28	β-Farnesene	1438	1.6
29	Geranyl acetone	1452	0.6
30	α-Caryophyllene	1456	0.7
31	*allo*-Aromadendren	1461	0.2
32	γ-Gurjunene	1473	0.9
33	α-Selinene	1495	1.1
34	β-Bisabolene	1507	1.3
35	Bisabolene	1522	0.7
36	1ξ,6ξ,7ξ-Cadina-4,9-diene	1502	0.8
37	γ-Cadinene	1512	1. 7
38	**Myristicin**	**1523**	**11.3**
39	Isopatchoulane	1552	1.9
40	Spathulenol	1578	1.3
41	Caryophyllene oxide	1584	1.6
42	*cis*-Farnesol	1697	1.0
43	*trans*-Farnesol	1724	0.7
	Total		94.7
	Monoterpenoids		60.5
	Sesquiterpenoids		21.3
	Others		12.9

Table 2: Toxicity of *Ostericum grosseserratum* essential oil against adult *Sitophilus zeamais*

Treatment	Contact toxicity			Fumigant toxicity		
	Mean LC_{50} (μg/adult) (95% CL)	***Slope ± SE***	***Chi square (χ^2)***	***Mean LC_{50} (mg/L air) (95% CL)***	***Slope ± SE***	***Chi square (χ^2)***
O. grosseserratum	17.97 (16.84-19.31)	3.32 ± 0.29	14.11	13.70 (12.85-14.75)	3.16 ± 0.21	9.46
Pyrethrum extract	4.29* (3.86-4.72)	0.72 ± 0.01	13.51	-	-	-
MeBr***	-	-	-	0.67**	-	-

*Data from Liu et al [11]; **data from Liu and Ho [10]; *** methyl bromide.*

against maize weevil but needs to be further evaluated for safety in humans and to enhance its activity.

ACKNOWLEDGEMENT

This work was funded by the Hi-Tech Research and Development of China 2011AA10A202 and National New-Drug Innovation Project 2009ZX09501-014. The authors thank Dr. QR Liu of the College of Life Sciences, Beijing Normal University, Beijing 100875, for the identification of the investigated plant.

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