Journal of Scientific Research & Reports
11(6): 1-6, 2016; Article no.JSRR.28523
ISSN: 2320-0227

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Volatile Constituents and Insecticidal Activity of Essential Oil of *Margaritaria discoidea* (Baill.) G. L. Webster

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Authors' contributions

This work was carried out in association with all authors. Authors SES and AAO collected the plant sample and performed the hydrodistillation of the oil sample while authors IAO and OAL designed the study and performed the analysis of the oil samples. Authors IAO and OIE managed the literature searches. Author IAO wrote the first and final draft of the manuscript. All authors read and approved the final manuscript.

Article Information

DOI: 10.9734/JSRR/2016/28523
Editor(s):
(1) Surapong Pinitglang, Department of Food Science and Technology, School of Science and Technology, University of the Thai Chamber of Commerce, Thailand.
Reviewers:
(1) Goudoum Augustin, The University of Maroua, Cameroon.
(2) Anonymous, Mercyhurst University, USA.
Complete Peer review History: http://www.sciencedomain.org/review-history/15873

Original Research Article

Received 23rd July 2016
Accepted 10th August 2016
Published 20th August 2016

ABSTRACT

The volatile constituents and insecticidal activity of essential oil obtained by hydrodistillation of air-dried and pulverized leaves of *Magaritaria discoidea* (Baill.) G. L. Webster growing in Nigeria was studied. The constituents of the oil were analysed by means of gas chromatography (GC) and gas chromatography coupled with mass spectrometry (GC-MS). The percentage yield of oil was 0.10% (v/w), calculated on a dry weight basis. A total of fifteen compounds, representing 99.99% of the total component of the essential oil of *M. discoidea* were characterized. The major compounds identified in the oil were geranyl acetone (33.0%), phytol (12.7%) and (*E*)-α-ionone (11.6%). The result of insecticidal activity indicated that *M. discoidea* essential oil at a concentration of 120 mg/mL displayed 100% mortality against *Sitophilus zeamais*, at 72 h. This is the first report on the

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chemical constituents and insecticidal activity of essential oil of *M. discoidea*.
Aims: The aim of this study was to isolate essential oils from the leaf of *Magaritaria discoidea* (Baill.) G. L. Webster (Euphorbiaceae) and investigate the volatile constituents present therein as well as insecticidal potential.
Study Design: The study involves the hydrodisitillation of essential oils from the air-dried plant materials, analysis of their chemical composition and evaluation of biological activity.
Place and Duration of Study: Fresh leaves of *M. discoidea* were harvested from trees growing at Tipper Garage, Kolobo, Abeokuta, Ogun State, Nigeria, in March 2016. The study was coonducted over 3 months period.
Methodology: About 500 g of air-dried plant samples was shredded and their oils were obtained by separate hydrodistillation for 4 h at normal pressure, according to the British Pharmacopoeia. The chemical constituents of the distilled oils were analyzed by means of gas chromatography-flame ionization detector (GC-FID) and gas chromatography coupled with mass spectrometry (GC-MS), while the insecticidal assay was conducted by established procedure.
Results: The main compounds of the leaf essential oil geranyl acetone (33.0%), phytol (12.7%) and (*E*)-α-ionone (11.6%). The result of biological activities indicated that *M. discoidea* essential, oil at a concentration of 120 mg/mL displayed 100% mortality against *Sitophilus zeamais*, at 72 h.
Conclusion: The present oil compositions were reported for the first for this species and for any member of *Magaritaria* family of plant.

Keywords: Margaritaria discoidea; Sitophilus zeamais; essential oil constituent; terpenes; insecticidal activity.

1. INTRODUCTION

Margaritaria discoidea (Baill.) G. L. Webster (Euphorbiaceae) is a deciduous shrub or tree with a spreading crown; usually growing up to 20 m tall. The bole is unbuttressed and can be 30cm in diameter. The flowers are fragrant and are much visited by bees and other insects [1]. The blue edible sub-globose fruit is up to 13 mm wide, containing a single large seed. The young leaves have occasionally been eaten. The extracts from various parts of *M. discoidea* showed significant antimicrobial, antioxidant and anti-inflammatory activities [2], displayed analgesic potential [3] and possesses gastroprotective activity [4]. A previous study observed that extracts and alkaloids of *M. discoidea* exhibited moderate antimicrobial, anti-plasmodia, anti-malarial and anti HIV activities [5]. The non-polar extracts of *M. discoidea* roots and/or leaves are potential sources of new microfilaricidal compounds for the treatment of onchocerciasis, *Onchocerca* microfilariae [6]. The organic extract of *M. discoidea* possessed stronger cytotoxicity on OVCAR-8, A2780 and cisplatin-resistant A2780 cis ovarian cancer cells [7]. A water soluble extract of *M. discoidea* induced high acaricidal activity in nymphs and adults of *Rhipicephalus appendiculatus,* and *Amblyomma variegatum* [8]. The stem bark of *M. discoidea* reportedly contains tannins which exerted anti-phlogistic activity through potent inhibition of cyclo-oxygenase-1 [9].

Ethyl esters of palmitic and linoleic acids were found as the major compounds in the GC–MS analysis of the hexane fraction of *M. discoidea* [4]. The flavonoids hydroxygenkwanin-8-C-[α-rhamnopyranosyl-(1→6)]-β-glucopyranoside, genkwanin-6-C-[α-rhamnopyranosyl(1→6)]-β-glucopyranoside, kaempferol-3-O-α-rhamnopyranosyl-(1→2)-β-glucopyranoside-7-O-α-rhamnopyranoside and kaempferol-3-O-α-rhamnopyranosyl-(1→2)-[α-rhamnopyranosyl-(1→6)]-β-glucopyranoside-7-O-α-rhamnopyranoside isolated from the stem bark of *M. discoidea* demonstrated free-radical scavenging and antibacterial activities [10]. The fractionation and purification of the leaf extract led to the isolation of *ent*-phyllanthidine, 14,15-dihydroallosecurinine-15-β-ol, securinine, securinol, and viroallosecurinine [6]. Gallic acid and securinine characterized from the plant were determined to contribute to the cytotoxic activity of the extract of *M. discoidea* [7]. Phylochrysine is a central nervous stimulant which may account in part for the plant's stimulatory properties [1].

Although, oil extract from the wood of this plant was found to have acaricidal activity [8]. However, to the best of our knowledge, no previous information concerning the composition of essential oil of *M. discoidea* was found in literature. The aim of the present study is to report the chemical constituents and insecticidal activity of the essential oil of *M. discoidea,* which has not been studied previously.

2. MATERIALS AND METHODS

2.1 Plant Material

Fresh leaves of *M. discoidea* were harvested from trees growing at Tipper Garage, Kolobo, Abeokuta, Ogun State, Nigeria, in March 2016. Botanical identification was carried out by Messrs Odewo A.O and Adeyemo A., at the Herbarium of Forestry Research Institute of Nigeria (FRIN), Ibadan, where a voucher specimen (FHI 110422) was deposited.

2.2 Hydrodistillation of Essential Oil

Briefly, 500 g of the pulverized sample were carefully introduced into a 5 L flask and distilled water (5 L) was added until it covers the sample completely. Hydrodistillation was carried out for 4 h in an all glass Clevenger-type distillation unit designed according to the specification [11]. The volatile oils distilled over water and were collected separately in the receiver arm of the apparatus into a clean and previously weighed sample bottles. The oils were kept under refrigeration until the moment of analyses.

2.3 Analysis of Essential Oil

2.3.1 Gas chromatography

Gas chromatography (GC) analysis of was carried out on a Hewlett Packard HP 6820 Gas Chromatograph equipped with a FID detector and DB-5 column (60 m x 0.25 mm id), film thickness was 0.25 µm and the split ratio was 1:25. The oven temperature was programmed from 50℃ (after 2 min) to 240℃ at 5℃/min and the final temperature was held for 10 min. Injection and detector temperatures were 200℃ and 240℃, respectively. Hydrogen was the carrier gas. An aliquot (0.5 µL of the diluted oil) was injected into the GC. Peaks were measured by electronic integration. A homologous series of *n*-alkanes were run under the same conditions for determination of retention indices.

2.3.2 Gas chromatography-mass spectrometry

Gas chromatography-mass spectrometry (GC-MS) analysis of the oil was performed on a Hewlett Packard Gas Chromatography HP 6890 interfaced with Hewlett Packard 5973 mass spectrometer system equipped with a DB-5 capillary column (30 m x 0.25 mm id, film thickness 0.25 µm). The oven temperature was programmed from 70-240℃ at the rate of 5℃/min. The ion source was set at 240℃ and electron ionization at 70eV. Helium was used as the carrier gas at a flow rate of 1 mL/min. Scanning range was 35 to 425 amu. The oil was diluted oil in *n*-hexane (1: 10) and 1.0 µL was injected into the GC/MS.

2.3.3 Identification of constituents of the oils

The identification of constituents was performed on the basis of retention indices (RI) determined by co-injection with reference to a homologous series of *n*-alkanes, under identical experimental conditions. Further identification was performed by comparison of their mass spectra with those from NIST [12] and the home-made MS library built up from pure substances and components of known essential oils, as well as by comparison of their retention indices with literature values [13].

2.4 Determination of Insecticidal Activity

The insecticidal activity was evaluated as described previously [14] using the maize weevil (*Sitophilus zeamais,* 3 months old). Different concentrations (40, 80, 120 and 150 mg/mL) of *M. discoidea* essential oil prepared separately and diluted in DMSO were tested on *S. zeamais*. The appropriate concentrations were applied to filter paper (Whatman number 1, cut into 7 cm diameter) and immediately introduced into Petri dish and sealed. For the control group, the insects were placed in the Petri dish under the same conditions but without the essential oil. Each concentration and control was replicated three times. Insect mortality was determined by observing the recovery of immobilized insects in 12 h intervals up to 72 h. When no movements were observed, insects were considered dead.

2.5 Statistical Analysis

The mean and standard deviation of three experiments were determined. Statistical analysis of the differences between mean values obtained for experimental groups were calculated using Microsoft excel program, 2003. Data were subjected to one way analysis of variance (ANOVA). P values ≤ 0.05 were regarded as significant and P values ≤ 0.01 as very significant. Mortality percentages were calculated by the correction formula for natural mortality in the untreated control [15]. The Lethal concentrations (LC_{50}) values for the insecticidal activity were calculated using probit analysis program, version 1.5.

3. RESULTS AND DISCUSSION

The percentage yields of volatile oil extract of *M. discoidea* was 0.10% (v/w), calculated on a dry weight basis. Table 1 indicates the chemical constituents present in the oils, their percentages as well as retention indices on DB-5 column. Fifteen compounds accounting for 99.9% of the total oil content were identified from the light yellow coloured oil. The main classes of compounds present in the leaf oil were oxygenated monoterpenes (54.1%), sesquiterpene hydrocarbons (12.6%) and oxygenated sesquiterpenes (10.0%) and diterpenes (12.7%).

The main constituents of the oil were geranyl acetone (33.0), (*E*)-α-ionone (11.6%9) and phytol (12.7%). There were significant quantity of δ-3-carene (7.4%), eremophilene (6.7%), caryophyllene oxide (4.3%), and α-ylangene (4.1%). The authors are unaware of any information on the chemical composition of essential oil of *M. discoidea*. The present study may represent the first of its kind aimed at the isolation and characterization of the volatile compounds of *M. discoidea* and in fact any *Margaritaria* species. The contents of geranylacetone, α-ionone and β-ionone, makes them useful in perfumery e.g soap perfumes, fragrance and flavouring.

The insecticidal potential of *M. discoidea* essential oil against the adults of *S. zeamais* is given in Fig. 1 and Table 2, respectively. The results demonstrated a clear dose dependent bioassay ($P < 0.05$), where the mortality was positively correlated with different concentrations of the oil. Fig. 1 show the percentage mortality of *M. discoidea* essential oil against the adults of *S. zeamais* at different concentrations. At a dose of 40 mg/mL, the mortality rate of *S. zeamais* by the oil of *M. discoidea* was 35.7% at 24 h of exposure, while at 72 h, mortality rate was 92.9%. However, at a dose of 120 mg/mL and 72 h of exposure, the oil has a mortality rate of 100% comparable to that of allethrin and permethrin. In addition, the result log-probit analysis (LC_{50}) with 95% confidence level of the oil (Table 2) progressively decreases as the time of exposure increases. Although *M. discoidea* essential oil has toxic effect on *S. zeamais* and comparable insecticidal activity to the permethrin, but, insecticidal effects of some compounds and/or groups of constituents present in *M. discoidea* essential oil were comparable to allethrin [15], advocating further screening of the oil.

Table 1. Chemical constituents of essential oils of *M. discoidea*

Compounds[a]	RI [b]	RI [c]	Percentage composition
Artemiseole	980	971	4.8
δ-3-Carene	1011	1008	7.4
1-Pentanol-5-(methylenecyclopropyl)	1092	1092	3.1
α-Ylangene	1377	1373	4.1
β-Ylangene	1420	1419	1.0
(*E*)-α-Ionone	1430	1428	11.6
Geranyl acetone	1456	1453	33.0
(*E*)-β-Ionone	1489	1487	4.7
Eremophilene	1498	1498	6.7
α-Muurolene	1501	1500	0.8
Caryophyllene oxide	1583	1582	4.3
trans, trans-Pseudoionone	1587	1584	2.0
α-Bisabolol	1690	1685	1.5
trans-Geranyl geraniol	2028	2026	2.2
Phytol	2125	2119	12.7
Total			99.9
Monoterpene hydrocarbons			7.4
Oxygenated monoterpenes			54.1
Sesquiterpene hydrocarbons			12.6
Oxygenated sesquiterpenes			10.0
Diterpenes			12.7
Non-terpenes			3.1

[a] Elution order on DB-5 column; [b] Retention indices on DB-5 column; [c] Literature retention indices (see experimental)

Table 2. Insecticidal activity of *M. discoidea* essential oil against *S. zeamais*[a]

	IC_{50} (95 CI)[b]		
	24[c]	**48**	**72**
*Md*EO[d]	62.69 (49.67 – 77.29)	32.49 (24.86- 41.12)	21.26 (13.71- 34.03)
Allethrin[e]	-	-	7.4 (2.01 – 14.65)
Permethrin[e]	-	-	11.10 (6.03 – 23.19)

[a](n= 3, X ± SEM; [b]LC_{50} - Lethal concentrations with 50 % mortality; [c] – Time; [d]MdEO – M. discoidea essential oil; [e] – Controls.

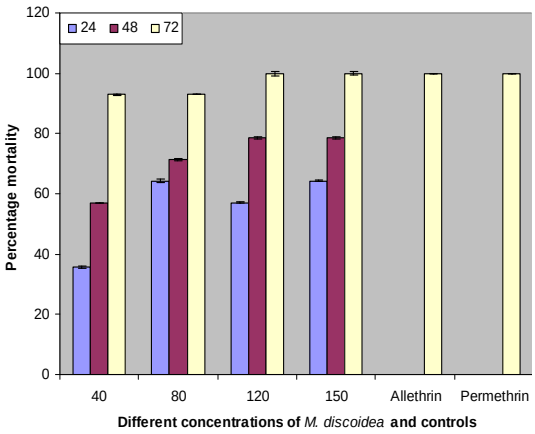

Fig. 1. Percentage mortality of *M. discoidea* essential oil against the adults of *S. zeamais*

It is well known that the biological activity of essential oils may depend on the major compound(s) and/or synergy between some major and minor compounds [16]. Essential oils containing a sizeable proportion of gernayl acetone have displayed larvicidal activity against mosquito [17,18], toxicity to housefly [19] and repellents to *Aedes aegypti* [20]. Plant extracts and essential oils containing large amount of phytol are known to exhibit insecticidal activity against *S. zeamais* [15,21] and some other insects such as sweet potato weevil, *Cylas formicarius elegantulus* [22], *Musca domestica* [23] and immature whitefly *Bemisia tabaci* [24].

4. CONCLUSION

In the present investigation of chemical constituents of essential oils from *M. discoidea,* geranyl acetone, phytol and (*E*)-α-ionone were the main compounds. The result of biological activities indicated that *M. discoidea* essential oil displayed mortality against *S. zeamais.* Due to the non-availability of literature citation, the results could not compare with previous study on the essential oil of the plant.

CONSENT

It is not applicable.

ETHICAL APPROVAL

It is not applicable.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

1. Burkil HM. The useful plants of West Tropical Africa. Royal Botanic Gardens: Kew. 1985 – 2004.
2. Dickson RA, Fleischer TC, Ekuadzi E, Mensah AY, Annan K, Woode E. Antibacterial, antioxidant and anti-inflammatory properties of *Margaritaria discoidea*, a wound healing remedy from Ghana. Phcog J. 2010;2(17):32-9.
3. Adedapo AA, Sofidiya MO, Afolayan AJ. Anti-inflammatory and analgesic activities of the aqueous extracts of *Margaritaria discoidea* (Euphorbiaceae) stem bark in experimental animal models. Rev Biol Trop. 2009;57(4):1193-200.
4. Sofidiya MO, Orisaremi CO, Sansaliyu I, Adetunde TO. Gastroprotective and antioxidant potentials of ethanolic stem bark extract of *Margaritaria discoidea* (Euphorbiaceae) in rats. J Ethno-pharmacol. 2015;171(2):240-246.
5. Diallo MST, Baldé MA, Camara A, Traoré MS, Bah ML, Diallo AS, et al. Ethnomedical, phytochemical and biological investigations of *Margaritaria discoidea* (Baill.) Webster, a plant species widely used in Guinean traditional medicine. J. Plant Sci. 2015;3(2):40-46.
6. Cho-Ngwa F, Abongwa M, Ngemenya MN, Nyongbela KD. Selective activity of extracts of *Margaritaria discoidea* and

Homalium africanum on *Onchocerca ochengi.* BMC Complem Alter Med BMC. 2010;10(62).
DOI: 10.1186/1472-6882.

7. Johnson-Ajinwo OR, Richardson A, Li WW. Cytotoxic effects of stem bark extracts and pure compounds from *Margaritaria discoidea* on human ovarian cancer cell lines. Phytomed. 2015;22(1):1-4.
8. Kaaya GP, Mwangi EN, Malonza MM. Acaricidal activity of *Margaritaria discoidea* plant extracts against the ticks *Rhipicephalus appendiculatus* and *Amblyomma varigatum*. Int J Acarol. 1995;21(2):123-9.
9. Xu GJ. Chinese medicine and technology people's medical publishing house. Beijing, China. 1996;1:46–50.
10. Ekuadzi E, Dickson R, Fleischer T, Annan K, Pistorius D, Oberer L, et al. Flavonoid glycosides from the stem bark of *Margaritaria discoidea* demonstrates antibacterial and free radical scavenging activities. Phytother Res. 2014;28(5):784-7.
11. British Pharmacopoeia. London: HM Stationary Office. 1980;2.
12. NIST. Chemistry Web Book. Data from NIST Standard Reference Database. 2011;69.
Available: http://www.nist.gov/
13. Joulain, Koenig. The atlas of spectral data of sesquiterpene hydrocarbons. E. B. Verlag, Hamburg; 1998.
14. Ilboudo Z, DabiréL CB, Nébié RCH, et al. Biological activity and persistence of four essential oils towards the main pest of stored cowpeas, *Callosobruchus maculatus* (F.) (Coleoptera: Bruchidae). J Stored Prod Res. 2010;46(2):124-8.
15. Abbott WS. A method for computing the effectiveness of an insecticide. J Econ Entom. 1925;18(3):265-267.
16. Lawal OA, Ogunwande IA, Salvador AF, Sanni AA, Opoku AR. *Pachira glabra* Pasq. Essential oil: Chemical constituents, antimicrobial and insecticidal activities. J Oleo Sci. 2014;63(6):629-35.
17. Oyewole IO, Moronkola DO, Ogunwande IA, Okoh H, Ibidapo CA, Denloye AAB, et al. Larvicidal activity of the essential oil from *Phyllanthus amarus* Sch. et Thonn (Euphorbiaceae) against three species of mosquitoes. Der Pharm Lett. 2010;2(6): 136-41.
18. Innocent E, Cosam CJ, Nicholas KG, Mayunga HHN, Ahmed H. Constituents of the essential oil of *Suregada zanzibariensis* leaves are repellent to the mosquito, *Anopheles gambiae* s.s. J Insect Sci. 2010;10(57).
DOI: insectscience.org/10.57.
19. Mann RS, Kaufman PE, Butler JF. Evaluation of semiochemical toxicity to houseflies and stable flies (Diptera: Muscidae). Pest Manag Sci. 2010;66(8): 816-24.
20. Jaramillo RGI, Logan JG, Loza-Reyes E, Stashenko E, Moores GD. Repellents Inhibit P450 Enzymes in *Stegomyia (Aedes) aegypti*. PLoS ONE. 2012;7(11): e48698.
DOI:10.1371/journal.pone.0048698
21. Lawal OA, Opoku RA. Ogunwande IA. Phytoconstituents and insecticidal activity of different leaf solvent extracts of *Chromolaena odorata* against *Sitophilus zeamais.* Eur J Med Pl. 2015;5(3):237-7.
22. McNeil MJ, Roy P, Williams LAD. Chemical composition and biological activity of the essential oil from Jamaican *Cleome serrata*. Nat Prod Comm. 2012;7(9):1231-2.
23. Chauhan N, Kumar P, Mishra S, Verma S, Malik A, Sharma S. Insecticidal activity of *Jatropha curcas* extracts against housefly, *Musca domestica*. Environ Sci Pollut Res Int. 2015;22(19):14793-800.
24. Cruz-Estrada A, Gamboa-Angulo M, Borges-Argáez R, Ruiz-Sánchez E. Insecticidal effects of plant extracts on immature whitefly *Bemisia tabaci* Genn. (Hemiptera: Aleyroideae). Elect J Biotech. 2013;16(1):1-6.

Peer-review history:
The peer review history for this paper can be accessed here:
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