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Mosquito Larvicidal Activity on *Aedes albopictus* and Constituents of Essential Oils from *Manglietia dandyi* (Gagnep.) Dandy

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Abstract: Herein, we report the results of our investigation on essential oils obtained by hydrodistilation of the leaves and fruits of *Manglietia dandyi* (Gagnep.) Dandy. The constituents of the oils were analyzed by gas chromatography-flame ionization detector (GC-FID) and gas chromatography-mass spectrometry (GC-MS). The yields of the oils were 0.25% and 0.21% (v/w, leaf and fruit respectively), calculated on a dry weight basis. The main constituents of the leaf oils were (*E*)-nerolidol (18.4%) and α -selinene (11.0%) while the fruit oils contained abundance of β -caryophylene (27.7%), δ -cadinene (13.7%), α -humulene (13.2%) and α -copaene (11.6%). The results indicated that *M. dandyi* leaf oil exhibited 96% and 100% mortality towards the fourth-instant larvae of *Aedes albopictus* at 24 h at tested concentrations of 50 µg/mL and 100 µg/mL. The oil only displayed 100% mortality at concentrations of 50 and 100 µg/mL and under 48 h. The minimum lethal concentrations, LC₅₀, were 29.57 µg/mL (24 h) and 29.02 µg/mL (48 h); while the LC₉₀ values of 46.21 µg/mL and 42.29 µg/mL were obtained respectively at 24 h and 48 h µg/mL. The chemical constituents and larvicidal action of *M. dandyi* essential oils are being reported for the first time.

Keywords: *Manglietia dandyi*; essential oil; sesquiterpenes; larvicidal activity. © 2020 ACG Publications. All rights reserved.

1. Plant Source

In the course of extensive studies on aromatic plants from Vietnam [1,2], we investigated *Manglietia dandyi* (Gagnep.) Dandy (Magnoliaceae). Herein, the chemical constituents of the essential oils from the leaf and

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fruit as well as the larvicidal activity of the leaf oil were reported. The leaves and fruits of *M. dandyi* were collected from Vũ Quang National Park, Hà Tĩnh (GPS: 18°20'N 105°54'E), Vietnam, in August 2018, and identified by Dr. Ngoc Dai. A voucher specimen (LDL 719) was deposited at the Botany Museum, Nghệ An College of Economics, Vietnam.

2. Previous Studies

Till moment no previous investigation has been carried out on the volatile composition and biological activity of both volatile and non-volatile components of *M. dandyi*. However, sesquiterpene compounds were reported to constitute the main constituents of other *Maglienta* oils analysed previously from Vietnam [3] and China [4].

3. Present Study

Hydrodistillation of the air dry parts of *M. dandyi* produced strong, odorous clear yellow oils. The volatile oils were obtained in yields of $0.25 \pm 0.01\%$ (v/w, leaf) and $0.21 \pm 0.01\%$ (v/w, fruit). Table 1 summarizes the identified components in order of their elution on the HP-5MS column used for the GC-MS analysis. Forty-seven compounds representing 93.8% of the oil contents were identified in the leaf of *M. dandyi*. These comprised of sesquiterpene hydrocarbons (49.9%), oxygenated sesquiterpenes (30.9%), monoterpene hydrocarbons and oxygenated monoterpenes (2.4%). The main constituents of the leaf oil were (*E*)-nerolidol (18.4%) and α -selinene (11.0%). In addition, significant quantity of δ -cadinene (7.5%), α -copaene (6.0%) and β -selinene (6.0%) were also present in the oil. On the other hand, 53 compounds accounting for 99.2% of the oil contents were identified in the fruit of *M. dandyi*. Sesquiterpene hydrocarbons (82.0%) along with oxygenated sesquiterpenes (8.4%), monoterpene hydrocarbons (4.8%) and oxygenated monoterpenes (4.0%) represent the classes of compounds identified in the fruit oil. β -Caryophylene (27.7%), δ -cadinene (13.7%), α -humulene (13.2%) and α -copaene (11.6%) were the compounds occurring in higher amounts in the fruit oil.

Compound	RI ^a	RI ^b	Leaf ^c	Fruit ^c
α-Pinene	937	937	0.7	0.7
Camphene	954	946	0.6	1.1
β-Pinene	983	980	1.0	0.2
Myrcene	990	988	0.5	0.2
α-Phellandrene	1009	1004	0.6	0.5
α-Terpinene	1020	1014	0.3	0.1
o-Cymene	1028	1026	2.4	0.5
Limonene	1032	1030	3.7	0.8
β-Phellandrene	1034	1032	0.8	0.4
γ-Terpinene	1062	1056	-	0.2
Terpinolene	1092	1093	-	0.1
Linalool	1102	1100	2.3	1.7
Borneol	1176	1168	-	0.3
α-Terpineol	1198	1197	0.1	-
Geraniol	1256	1254	-	0.3
Bornyl acetate	1292	1297	-	0.7
α-Cubebene	1358	1356	-	0.2
Cyclosativene	1380	1371	-	0.1
Isoledene	1380	1373	-	0.1
Geranyl acetate	1382	1380	-	1.0
α-Ylangene	1383	1383	-	0.2
α-Copaene	1388	1387	6.0	11.6
α-Gurjunene	1423	1420	1.0	0.5
-				

Table 1. Chemical composition of essential oils of Manglietia dandyi

Table 1 continued				
β-Caryophylene	1437	1432	2.6	27.7
β-Gurjunene	1443	1438	0.2	0.5
Aromadendrene	1445	1440	2.2	1.2
(Z) - β -Farnesene	1458	1447	0.7	0.1
α-Humulene	1471	1465	1.2	13.2
9- <i>epi</i> -(<i>E</i>)-Caryophyllene	1477	1470	0.5	0.5
trans-Cadina-1(6),4-diene	1486	1484	-	1.4
β-Chamigrene	1487	1487	1.8	-
γ-Muurolene	1488	1488	1.8	1.3
α-Amorphene	1492	1490	0.5	0.2
β-Selinene	1503	1502	6.0	1.1
γ-Amorphene	1509	1510	0.2	3.7
α-Selinene	1511	1513	11.0	-
γ-Cadinene	1528	1525	1.1	0.7
δ-Cadinene	1532	1530	7.5	13.7
<i>cis</i> -Calamenene	1537	1534	2.4	1.0
Zonarene	1539	1540	1.0	0.7
trans-Cadina-1,4-diene	1546	1544	_	0.9
α-Cadinene	1552	1548	0.4	0.3
α-Calacorene	1558	1554	0.7	0.9
Elemicine	1559	1556	0.7	-
(E)-Nerolidol	1568	1560	18.4	1.3
β-Calacorene	1578	1578	-	0.2
Caryophyllene alcohol	1591	1588	0.3	0.2
Viriflorol	1603	1600	3.6	1.9
Cubeban-11-ol	1612	1606	0.8	0.3
Rosifoliol	1621	1620	0.6	-
Copaborneol	1625	1628	-	0.7
Humulene epoxide II	1630	1632	-	0.2
α-Corocalene	1637	1637	0.2	-
5-Guaiene-11-ol	1641	1640	0.9	0.2
1-epi-Cubenol	1645	1644	0.8	0.8
Cubenol	1651	1652	0.4	-
<i>epi</i> -α-Cadinol	1659	1660	0.6	1.1
α-Muurolol	1662	1664	1.0	0.4
α-Cadinol	1672	1666	0.8	0.9
neo-Intermedeol	1676	1672	2.3	0.4
Cadalene	1693	1690	0.2	-
(E,E)-Farnesol	1726	1722	0.4	-
		Total	93.8	99.2
Hydrocarbons			10.6	4.8
Oxygenated monoterpenes			2.4	4.0
Sesquiterpene hydrocarbons			49.9	82.0
Oxygenatedsesquiterpenes			8.4	

^a Retention indices on HP-5MS column; ^b Literature retention indices [5]; ^c Standard deviation were insignificant and excluded from the Table to avoid congestion; (-) Not identified

The high content of sesquiterpene compounds in the studied M. dandyi oils was in agreement with findings [3, 4]. This class of compounds occurred in abundance quantity in *Maglienta* oils analysed previously from Vietnam [3] and other parts of the world [4]. For example, the leaf oil of *M. fordiana* contained δ -cadinene (18.0%), (*E*)-nerolidol (16.7%), α -copaene (12.8%) and α -selinene (9.1%) while β -caryophyllene (29.9%), α -humulene (7.4%), (E)-nerolidol (6.3%) were the main compounds in *M. conifera* [3]. High contents of (*E*)-

nerolidol (14.61%) and δ -cadinol (20.57%) were identified in *M. moto*, while (*E*)-nerolidol (11.92%) and δ -cadinene (10.84%) were present in *M. yuyanensis* growing in China [4].

The mosquito larvicidal activity of *M. dandyi* leaf oil depicted by percentage mortality and lethal concentrations are summarized in Table 2. The oil sample exhibited 96% (concentration of 50 µg/mL) and 100% (concentration of 100 µg/mL) mortality against the larvae of *Ae. albopictus* at 24 h. Likewise, *M. dandyi* displayed 100% mortality against the larvae of *Ae. albopictus* at 48 h (concentrations of 50 and 100 µg/mL). There was no mortality in the EtOH used as control. The percentage mortality was dependent on the concentration of the tested oil samples. Thus, higher inhibition of mosquito larvae was observed in the 100 mg/L compared to other concentrations. From Table 2, the *M. dandyi* leaf oil exhibited larvicidal action towards *Ae. albopictus* with LC₅₀ values of 29.57 µg/mL and LC₉₀ 46.21 µg/mL at 24 h while LC₅₀ values of 29.02 µg/mL and LC₉₀ of 42.29 µg/mL were obtained at 48 h. Permethrin, the standard drug used as control displayed larvicidal activity at much lower values. These findings showed that the concentrations of test substances affected degree of toxicity and mortality rates. The larvicidal activity of *Maglienta* oil samples has not been the subject of much literature discussion. Overall results in this study showed that essential oil of *M. dandyi* exhibited good mortality and larvicidal activity on *Ae. Albopictus* larvae when compared to *M. garrettii* [6]. Also, *Magnolia grandiflora* of the Magnoliaceae family displayed larvicidal activity against *Ae. aegypti* with LD₅₀ values in the range between 51.5 and 54.7 ppm [7].

	Mortality (%)			
Concentration (µg/mL)	24 h ^a	48 h ^a		
12.5	5 ±0.000	10 ±0.000		
25	15±1.633	27.5 ± 1.915		
50	96 ± 0.500	100 ± 0.000		
100	100 ± 0.000	100 ± 0.000		
Minimum lethal concentration	(µg/mL) ^b			
LC_{50}^{c}	29.57 (25.940-34.766)	29.02 (25.836-36.496)		
LC_{90}^{d}	46.21 (36.103-55.434)	42.29 (38.594-62.060)		
LC _{50(permethrin)}	1.32			
LC ₉₀ (permethrin)	1.01			

Table 2. Larvicidal activity of Manglieta dandyi leaf essential oil against Aedes albopictus

^a (n= 4), There was no mortality in the EtOH controls; ^b values in brackets are 95% confidence level; Linear Regression Equations: ${}^{c}y = -2.277 + 0.077x$, $[X^{2} (5.250), P<0.001]$; ${}^{d}y = -2.804 + 0.097x$, $[X^{2} (5.407), P<0.001]$.

The larvicidal action of *M. dandyi* was comparable to many other essential oils screened against *Ae. albopictus.* The minimum lethal concentrations (LC₅₀) of *Ruta chalepensis,* Artemisia *verlotiorum* and *L*avandula *dentata* essential oils against the vector mosquito of *Ae. albopictus,* were 93.60, 324.00 and 602.80 µL/L respectively [8]. The essential oils of *Erechtites hieraciifolius* and *E. valerianifolius* showed good larvicidal activity against *Ae. albopictus* with LC₅₀ 10.5 and 5.8 µg/mL, respectively at 24 h test period [9]. *Eucaluptus nitens* exhibited larvicidal action against *Ae. albopictus* with LC ₅₀ of 28.19 ppm [10], while LC₅₀ value of 72.86 µg/mL was obtained for *A. macrostemon* [11]. Previous study revealed that the essential oil of *Severinia monophylla* showed potent larvicidal activity against *Ae. albopictus* with LC₅₀ value of 36 µg/mL after 48 h [12]. *Eucalyptus globulus* and *Azadirachta indica* were also shown to be effective larvicides with LC₅₀ of 93.3 ppm and 78.52 ppm respectively [13]. The essential oils of *Amyris balsamifera*, *Piper nigrum, Cinnamonum zeylanicum, Anethum graveolens, Jasminum grandiflorum, Juniperus communis and Thymus serpyllum* displayed larvicidal activity against *Ae. albopictus* with LC₅₀ values of 39.8, 62.78, 45.58, 10.52, 49.99, 53.22 and 60.31 ppm respectively [14]. The oil of *Toddalia asiatica* exhibited toxicity towards *Ae. albopictus* with LC₅₀ of 69.09 µg/mL [15].

The observed mosquito larvicidal activity of the essential oil may be due to actions of the major compounds or a synergy between the major and some minor constituents. The larvicidal actions of the main compounds of the essential oils have been reported. (*E*)-Nerolidol, a main constituent of the essential is known for its insecticidal and ovipositor deterrent [16,17]. Essential oil with high contents of (*E*)-nerolidol and α -selinene displayed toxicity towards larvae of *Ae. albopictus* [16]. A mixture of (*E*)-caryophyllene, α -humulene and terpinen-4-ol efficiently deterred oviposition in insects [18]. Cadinene, amongst others displayed the most effective larvicidal activity against *Anopheles stephensi*, *Ae. aegypti* and *Culex quinquefasciatus* [19].

Conclusions

The current study, being the first of its kind, provided information on the medicinal uses and phytochemical compounds in the essential oils of the leaves and fruits of *M. dandyi*. The major compounds were identified as *(E)*-nerolidol, α -selinene, β -caryophylene, δ -cadinene, α -humulene and α -copaene. In addition, the leaf oil fraction of *M. dandyi* displayed potent mortality and larvicidal activity against *Ae. albopictus*

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Supporting Information

Supporting Information accompanies this paper on <u>http://www.acgpubs.org/journal/records-of-natural-products</u>

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