

Annonaceae Essential Oils: Antimicrobial and Compositions of the Leaves of *Uvaria hamiltonii* Hook. f. & Thoms. and *Fissistigma kwangsiensis* Tsiang & P. T. Li

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Abstract: In this study, essential oils hydrodistilled from the leaves of *Uvaria hamiltonii* Hook. f. & Thoms. and *Fissistigma kwangsiensis* Tsiang & P. T. Li (Annonaceae) collected in Vietnam were analyzed by gas chromatographic techniques, and screened for the antimicrobial activities. The composition of both essential oils was dominated by sesquiterpenoids (70.0% and 78.7%, respectively). The main constituents of *U. hamiltonii* were germacrene D (22.9%), β -caryophyllene (21.1%), bicyclogermacrene (11.2%) and caryophyllene oxide (8.6%). The essential oil of *F. kwangsiensis* also showed abundant sesquiterpenes β -caryophyllene (24.5%), δ -cadinene (13.4%) and α -copaene (5.6%), but also included (*Z*)- β -ocimene (6.7%). The leaf oil of *U. hamiltonii* demonstrated notable antimicrobial activity against *Enterococcus faecalis* ATCC299212 with minimum inhibitory concentration (MIC) value of 7.99 μ g/mL and *Bacillus cereus* ATCC14579 (MIC 5.67 μ g/mL) while *F. kwangsiensis* showed the most potent activity towards *Pseudomonas aeruginosa* ATCC27853 and *Candida albicans* ATCC10231, with MIC values of 3.45 μ g/mL and 16.45 μ g/mL, respectively. Both essential oils should be considered for further investigation as renewable “green” antimicrobial agents.

Keywords: Antimicrobial activity; essential oil composition; sesquiterpenes. © 2021 ACG Publications. All rights reserved.

1. Plant Source

The genus *Uvaria* and *Fissistigma* are important members of the family Annonaceae. *Uvaria hamiltonii* Hook. f. & Thoms. and *Fissistigma kwangsiensis* Tsiang & P. T. Li are newly recorded flora of

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Constituent and antimicrobial activity of *U. hamiltonii* and *F. kwangsiense* leaf essential oil

Vietnam [1,2]. In the present communication, essential oils were obtained by hydrodistillation of the leaves of *U. hamiltonii* and *F. kwangsiensis* from its natural habitat in Vietnam, then analyzed by gas chromatography (GC) and gas chromatography-mass spectrometry (GC/MS) techniques, and subsequently screened for *in vitro* antimicrobial activities.

2. Previous Studies

The essential oil compositions of *U. grandiflora* [3], *U. microcarpa* [3], *U. rufa* [4] and *U. cordata* [4] from Vietnam; *U. chamae* [5] from Nigeria; *U. ovata* [6], *U. anonoides* [6] and *U. tortilis* [6] from Ivory Coast, were reported previously. In addition, *F. bracteolatum* [7], *F. villosissimum* [7], *F. latifolium* [7], *F. glaucescens* [7], *F. chloroneurum* [8], *F. cupreonitens* [8], *F. pallens* [8], *F. bicolor* [8], *F. shangtzeense* [8], *F. petelotii* [8], *F. maclurei* [9], *F. scandens* [10], *F. poilanei* [10] and *F. acumunatissimum* [10] essential oil compositions from Vietnam, as well as *F. shangtzeense* [11] from China, have also been reported. Sesquiterpene compounds were identified from the flower essential oil of *U. hamiltonii* [12]. To the best of our knowledge, this is the first report on the chemical composition of *U. hamiltonii* and *F. kwangsiensis* leaf essential oils from Vietnam, and the *in vitro* antimicrobial activity of these Vietnamese Annonaceae plants.

3. Present Study

The average yields of the essential oils were 0.25% and 0.31% ($\pm 0.01\%$, v/w), respectively for *U. hamiltonii* and *F. kwangsiensis*. Both essential oils were light yellow coloured. Thirty-five and fifty-one constituents accounting for 94.8% and 98.7% of the volatile contents were identified in the essential oils of *U. hamiltonii* and *F. kwangsiensis*, respectively, as seen in Table 1. The main classes of compounds present therein includes monoterpene hydrocarbons (5.3% and 10.0%), sesquiterpene hydrocarbons (70.0% and 78.7%) and oxygenated sesquiterpenes (19.0% and 10.0%) respectively. The oxygenated monoterpenes were not identified in *F. kwangsiensis* and present in lower quantity (0.5%) in *U. hamiltonii*. The major compounds of *U. hamiltonii* essential oil were germacrene D (22.9%), β -caryophyllene (21.1%), bicyclogermacrene (11.2%) and caryophyllene oxide (8.6%). The compositional pattern of *U. hamiltonii* essential oil was similar to *U. ovata* from Ivory coast [6] due to its high contents of germacrene D and β -caryophyllene. It should be noted that *Uvaria* essential exhibits chemical variability. For examples, monoterpenes were the dominant compounds of the leaf oils of *U. rufa* [4] and *U. grandiflora* [3], while fatty acids predominate in *U. cordata* [4], with phenylpropanoids making up the compositions of *U. microcarpa* [3]. The leaf oil of *U. anonoides* [6] was rich in sesquiterpenes and aromatic compounds. Sesquiterpene compounds such as γ -muurolene, cadina-1(10),4-diene, 7-*epi*- α -eudesmol, ylangene, aristolochene and longifolene were detected in the endogenous scent flower of *U. hamiltonii* [12]. Except γ -muurolene, all other compounds were not identified in the leaf oil under investigation.

On the other hand, β -caryophyllene (24.5%), δ -cadinene (13.4%) and (*Z*)- β -ocimene (6.7%) were the principal components of *F. kwangsiensis* oil. Terpenes were also the main constituents of majority of *Fissistigma* essential oils reported so far in the literature. These includes *F. chloroneurum* [8], *F. cupreonitens* [8], *F. pallens* [8], *F. bicolor* [8], *F. shangtzeense* [8], *F. petelotii* [8] and *F. maclurei* [9] where sesquiterpenes predominate. However, *F. scandens* [10], *F. poilanei* [10] and *F. acumunatissimum* [10] and *F. shangtzeense* [11] were dominated by monoterpene compounds. The identities of these terpene compounds were different from one species to another. A noteworthy observation is the fact that β -caryophyllene was the one of the main constituents of *F. cupreonitens* [8] and *F. bicolor* [8]. The existence of chemical variability in the essential oil compositions of *Fissistigma* species were previously reported [7-10].

Table 1. Constituents of the leaf essential oils of *U. hamiltonii* and *F. kwangsiensis*

Compound ^a	RI (Exp.)	RI ^[14]	<i>U. hamiltonii</i> ^b	<i>F. kwangsiensis</i> ^b
α -Thujene	930	921	-	0.2
α -Pinene	937	932	3.3	0.7
β -Pinene	983	978	0.5	1.0
Myrcene	990	988	-	0.3
o-Cymene	1029	1017	0.5	0.2
β -Phellandrene	1032	1026	0.2	0.1
Limonene	1034	1028	0.8	0.1
1,8-Cineole	1037	1032	0.5	-
(Z)- β -Ocimene	1038	1044	-	6.7
(E)- β -Ocimene	1049	1052	-	0.7
δ -Elemene	1347	1351	1.1	0.9
α -Cubebene	1359	1353	0.2	2.4
α -Ylangene	1384	1383	-	0.4
α -Copaene	1388	1389	1.6	5.6
β -Cubebene	1401	1405	-	1.2
<i>cis</i> - β -Elemene	1404	1407	3.1	-
β -Caryophyllene	1437	1437	21.1	24.5
β -Gurjunene	1444	1446	0.4	0.6
Aromadendrene	1455	1461	0.3	
Guaine-6,9-diene	1456	1463	-	0.5
α -Humulene	1471	1478	4.1	3.6
9- <i>epi</i> -(E)-Caryophyllene	1477	1479	-	0.5
<i>trans</i> -Cadinane-1(6),4-diene	1487	1487	-	1.2
<i>epi</i> -Zonarene	1491	1489	-	2.9
γ -Murolene	1490	1491	0.8	4.5
α -Amorphene	1493	1493	0.5	0.9
Germacrene D	1498	1499	22.9	1.9
β -Selinene	1503	1501	0.3	0.9
δ -Selinene	1504	1507	-	0.9
<i>trans</i> -Murolane-4(14),5-diene	1509	1512	0.3	1.0
α -Murolene	1512	1513	-	3.6
Bicyclogermacrene	1513	1515	11.2	-
δ -Amorphene	1520	1523	-	0.4
γ -Cadinene	1528	1534	0.5	2.2
δ -Cadinene	1532	1536	1.0	13.4
Zonarene	1540	1544	-	0.9
<i>trans</i> -Cadinane-1,4-diene	1547	1550	-	1.2
α -Cadinene	1552	1552	-	1.1
α -Calacorene	1558	1560	-	1.0
Germacrene B	1576	1578	0.6	0.3
β -Calacorene	1579	1581	-	0.2
Mintoxide	1585	1589	0.4	-
Spathulenol	1595	1596	5.8	0.4
Gleenol	1598	1603	-	0.2
Caryophyllene oxide	1605	1610	8.6	4.1
Guaiol (=Champacol)	1614	1616	-	0.3
Humulene epoxide II	1634	1635	1.1	0.6
α -Corocalen	1637	1640	-	0.6
1- <i>epi</i> -Cubenol	1644	1646	1.4	1.0
γ -Eudesmol	1649	1650	-	0.5
<i>epi</i> - α -Cadinol	1656	1656	0.2	0.5
<i>epi</i> - α -Murolol	1658	1660	0.5	-
δ -Cadinol	1660	1662	-	0.4
β -Eudesmol	1670	1676	-	0.6

Constituent and antimicrobial activity of *U. hamiltonii* and *F. kwangsiense* leaf essential oil

Table 1 continued..

α -Cadinol	1671	1678	0.3	-
α -Eudesmol	1672	1680	-	0.6
α -Asarone	1684	1692	0.4	-
<i>trans</i> -Calamene-10-ol	1686	1693	-	0.2
Eudesma-4(15),7-dien-1 β -ol	1704	1710	0.3	-
Total			94.8	98.7
Monoterpene hydrocarbons (Sr. No. 1-7, 9,10)			5.3	10.0
Oxygenated monoterpenes (Sr. No. 8)			0.5	-
Sesquiterpene hydrocarbons (Sr. No. 11-41)			70.0	78.7
Oxygenatedsesquiterpenes (Sr. No. 42-59)			19.0	10.0

^a Elution order on HP-5MS column; RI (Exp.) Retention indices on HP-5MS column; RI (Lit) Literature retention indices on HP-5MS column NIST [14]; ^b Standard deviation were insignificant and excluded

Antimicrobial Activity of the Essential Oils: The results of the antimicrobial test on *U. hamiltonii* and *F. kwangsiensis* essential oils are shown in Table 2. The essential oils displayed antimicrobial activity against four of the tested microorganisms and anti-candidal activity, with the minimum inhibitory concentration (MIC) values in most cases < 50 $\mu\text{g/mL}$. The leaf essential oil of *U. hamiltonii* was the most active towards *Enterococcus faecalis* ATCC299212 (MIC, 7.99 $\mu\text{g/mL}$), *Staphylococcus aureus* ATCC25923 (MIC, 20.34 $\mu\text{g/mL}$) and *Bacillus cereus* ATCC14579 (MIC, 20.34 $\mu\text{g/mL}$). However, *F. kwangsiensis* essential oil displayed the most potent antibacterial action and anti-candidal activity against *Pseudomonas aeruginosa* ATCC27853 (MIC, 3.45 $\mu\text{g/mL}$) and *Candida albicans* ATCC10231 (MIC, 16.45 $\mu\text{g/mL}$), respectively. The IC₅₀ values are normally in the range 10 -220 $\mu\text{g/mL}$. The obtained MIC and IC₅₀ values are indication that the leaf essential oils of *U. hamiltonii* and *F. kwangsiensis* exhibited potent antimicrobial and anti-candidal activities against the tested microorganisms. The essential oils, however, did not inhibit the growth of *Salmonella enterica* ATCC13076 and *Escherichia coli* ATCC25922 [15-17] (see supporting information for details).

Table 2. Antimicrobial activity of the leaf essential oils of *U. hamiltonii* and *F. kwangsiensis*

Microorganisms	MIC ($\mu\text{g/mL}$) ^a		IC ₅₀ ($\mu\text{g/mL}$) ^a	
	<i>U. hamiltonii</i>	<i>F. kwangsiensis</i>	<i>U. hamiltonii</i>	<i>F.</i>
<i>kwangsiensis</i>				
<i>Enterococcus faecalis</i> ATCC299212	7.99 \pm 0.01 ^b	33.62 \pm 0.50 ^c	16.0 \pm 0.11	64.0 \pm 0.10
<i>Staphylococcus aureus</i> ATCC25923	20.34 \pm 0.14 ^d	55.67 \pm 0.11 ^e	64.0 \pm 0.50	128.0 \pm 1.50
<i>Bacillus cereus</i> ATCC14579	5.67 \pm 0.50 ^f	12.45 \pm 0.50 ^g	16.0 \pm 0.50	32.0 \pm 0.50
<i>Pseudomonas aeruginosa</i> ATCC27853	12.34 \pm 0.50 ^h	3.45 \pm 0.00 ^h	32.0 \pm 0.50	16.0 \pm 0.15
<i>Candida albicans</i> ATCC10231	32.57 \pm 0.21 ^{&}	16.45 \pm 0.50 ^{&}	64.0 \pm 0.10	32.0 \pm 0.10

^a Mean value of three replicate assays; ^{b,c,d,e} Streptomycin, MIC values of 0.48 $\mu\text{g/mL}$, 2.07 $\mu\text{g/mL}$, 1.07, 3.20 $\mu\text{g/mL}$, 0.28 $\mu\text{g/mL}$, and 0.97 $\mu\text{g/mL}$, respectively; ^h Nystatine, MIC value of 8.0 $\mu\text{g/mL}$; [&] Cycloheximide, MIC of 3.20 $\mu\text{g/mL}$

Previous studies have shown that the biological activities of essential oils from different species of plants are dependent of the major compounds of abundance. In some other cases, synergies between the main and minor constituents have also enhanced the activity of natural products including essential oils [15,16]. These compounds exhibit activity by firstly destroy the microbial cytoplasmic wall to enhance permeability and passage of large protons and ions [17]. Nevertheless, the antibacterial effect can be sum up as cumulative actions of several compounds and not to a specific compound [17]. The sesquiterpene, β -caryophyllene showed antibacterial activity against *S. aureus* [18] and antifungal effect [19]. The composition pattern of essential oils is so complex and has made it difficult to explain the mode of antimicrobial action of particular oil. In addition, the variety of compounds of an essential oil serves as positive factor that may limit the development of resistance which is otherwise very common for synthetic drug [20]. Thus, essential oils of *U. hamiltonii* and *F. kwangsiensis* and their constituents may be considered among the most promising alternative to synthetic chemicals.

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In conclusion, the essential oil compositions of *U. hamiltonii* and *F. kwangsiensis* are reported for the first time. *Uvaria hamiltonii* and *F. kwangsiensis* leaf essential oil showed notable antimicrobial and anti-candidal activities, and may be useful sources of natural botanical agents.

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Supporting Information

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

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