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Leaf Essential Oil Composition of Six *Syzygium* Species from the Western Ghats, South India

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Abstract: The *Syzygium* (Family: Myrtaceae) species are well known for their aromatic nature. Though 45 *Syzygium* species are reported from the Western Ghats region of India, the volatile oil chemistry of most of these aromatic plants are uninvestigated. The present study reports the chemical constituents of the leaf essential oils of 6 *Syzygium* species, *S. arnottianum* Walp., *S. caryophyllatum* (L.) Alston, *S. hemisphericum* (Wight) Alston, *S. laetum* (Buch. Ham.) Gandhi, *S. lanceolatum* (Lam.) Wight & Arn. and *S. zeylanicum* (L.) DC. var. *zeylanicum*, collected from the Western Ghats of Kerala. Sesquiterpenoids were the predominant compounds in all the *Syzygium* species studied and -caryophyllene and caryophyllene oxide were present in all the oils except *S. laetum.* The open chain sesquiterpenoids (*Z,E)*-α-farnesene and (*E)*-nerolidol were characteristic of *S. laetum* while phenyl propanoids were exclusively present in *S. lanceolatum.*

Keywords: *Essential Oil; GC-MS; Syzygium arnottianum; Syzygium caryophyllatum; Syzygium hemisphericum; Syzygium laetum; Syzygium lanceolatum; Syzygium zeylanicum.* © 2015 ACG Publications. All rights reserved.

1. Plant Source

Fresh leaves of the *Syzygium* species were collected from the forests of southern Western Ghats, Kerala, India and voucher herbarium specimens (TBGT No.) of *Syzygium arnottianum* Walp. (66407), *Syzygium caryophyllatum* (L.) Alston (50993), *Syzygium hemisphericum* (Wight) Alston (50959), *Syzygium laetum* (Buch. Ham.) Gandhi (66409), *Syzygium lanceolatum* (Lam.) Wight & Arn.(50992) and *Syzygium zeylanicum* (L.) DC. var. *zeylanicum* (50995) were deposited at the JNTBGRI herbarium (TBGT). The plant materials were identified by Dr. T. G. Vinodkumar, St. Thomas College, Ranni, Kerala.

2. Previous Studies

The genus *Syzygium* Gaertner (Family: Myrtaceae) is represented by nearly 1200 species in the old world tropics and 45species are reported in the Western Ghats of India [1]. Literature searches showed that there are no previous studies on the volatile constituents of the six *Syzygium* species

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reported here, while a few species such as *S. aromaticum* [2,3], *S. cumini* [4,5,6], *S. guineense* [7], *S. cordatum* [8], *S. gardneri* [9] and *S. semarangense* [10] were investigated for their leaf volatile

chemical constituents. Though *S. caryophyllatum* (L.) Alston is different from *S. aromaticum* (L.) Merr. & Perry (common clove tree), the chemistry of *S. aromaticum* has been reported under the title of *S. caryophyllatum* (L.) Alston [3].

3. Present Study

Isolation of Essential Oil: The essential oils were isolated by hydrodistillation of the fresh leaves (300g each) for 3 h. using a Clevenger type apparatus. The oils were dried over anhydrous sodium sulphate and stored at 4^{o}C until the analyses.

Analysis of Essential Oil: The GC-FID analysis was done on a Varian CP-3800 gas chromatograph fitted with CP Sil 8CB fused silica capillary column (30 m, 0.32 mm i.d., film thickness 0.25 μm) with FID detector using nitrogen as a carrier gas at flow rate of 1mL/ min. The split ratio was 1:40, and 0.1μL oil sample (1:5 dilution in diethyl ether) was injected. Oven temperature programme: injector temperature 220^{o}C, oven temperature 50-230^{o}C at 3^{o}C/ min., detector temperature 250^{o}C. Relative percentage of components was obtained from the peak area of volatiles. The GC/MS analysis was done on a Hewlett Packard 6890 gas chromatograph fitted with a cross-linked 5% PH ME siloxane HP-5 MS capillary column (30 m x 0.32 mm, film thickness 0.25 μm) coupled with a 5973 series mass selective detector under the following conditions with splitless injection of 1.0 μL of essential oil (1:10 dilution in diethyl ether), helium as the carrier gas at 1.4 mL/ min constant flow mode. The temperature programme for the analysis of the oils were, injector temperature 220^{o}C, oven temperature 60^{o}C to 246^{o}C (3^{o}C/min) and interface temperature 290^{o}C. Mass spectra: Electron Impact (EI^{+}) mode, 70 eV and ion source temperature 250^{o}C. The essential oil components were identified based on by MS library search (Wiley 2.75), relative retention indices calculated with respect to homologous of n-alkanes (C_6-C_{30}, Aldrich Chem. Co. Inc.) [11] and by literature reference [12].

The leaf essential oil yield (%v/w) was higher for *S. zeylanicum* (0.33%), followed by *S. hemisphericum* (0.17%), *S. arnottianum* (0.12%) and *S. lanceolatum* (0.10%), while the yield was negligible for *S. laetum* (0.01%) and *S. caryophyllatum* (0.01%).The major volatile constituents identified from the leaf essential oil of *Syzygium* species were caryophyllene oxide (15.4%) and selina-11-en-4α-ol (13.0%) for *S. arnottianum,* β-caryophyllene (32.4%), 1-epi-cubenol (11.8%) and δ-cadinene (10.0%) for *S. caryophyllatum,* β-caryophyllene (40.5%) and α-humulene (39.7%) for *S. hemisphericum,* (*Z,E)*-α-farnesene (21.5%), γ-amorphene (12.1%) and epi-α-cadinol (10.2%) for *S. laetum,* α-humulene (23.1%), β-caryophyllene (16.1%) and phenyl propanal (13.5%)for *S. lanceolatum,* and β-caryophyllene (11.1%), α-cadinol (12.2%), humulene epoxide II (17.6%) caryophyllene oxide (18.9%) and α-humulene (24.0%) for *S. zeylanicum.*

Sesquiterpenoids were the predominant compounds in all the *Syzygium* species studied (Table 1). Among the sesquiterpenoids, caryophyllene or its derivatives were detected in all the *Syzygium* species. In *S. hemisphericum*, 84.4% of the volatile constituents were caryophyllene analogues, followed by *S. zeylanicum* (71.6%), *S. lanceolatum* (54.2%), *S. caryophyllatum* (44.3%), *S. arnottianum* (20.2%) and *S. laetum* (2.8%). β-Caryophyllene and caryophyllene oxide were present in all the oils except *S. laetum.* α-Humulene was a predominant constituents in all the oils except *S. arnottianum* and *S. laetum.* Selinene and derivatives constituting 36.2% were predominant in *S. arnottianum* while the open chain sesquiterpenoids (*Z,E)*-α-farnesene and (*E)*-nerolidol were characteristic of *S. laetum*. Phenyl propanoids were exclusively present (14.4%) in *S. lanceolatum* and phenyl propanal can be considered as the chemotaxonomic marker compound for *S. lanceolatum*. Monoterpenoids were present in negligible amount only in *S. hemisphericum*(0.2%) and *S. zeylanicum* (0.9%).

Table 1.Essential oil constituents of the leaves of *Syzygium* species.

Compound	RRI	*S.arn*	*S.car*	*S.hem*	*S.lat*	*S.lan*	*S.zey*
β-Pinene	969	-	-	0.2	-	-	-
(Z)-β-Ocimene	1032	-	-	-	-	-	0.4
Linalool	1093	-	-	-	-	-	0.5
Phenyl ethyl alcohol	1100	-	-	-	-	0.9	-
Phenyl propanal	1156	-	-	-	-	13.5	-
δ-Elemene	1332	-	-	-	-	0.3	-
α-Copaene	1368	-	5.8	-	-	-	1.3
β-Bourbonene	1381	1.0	-	-	-	-	-
β-Elemene	1383	3.0	-	-	-	5.6	-
α-Gurjunene	1400	-	-	-	-	0.6	-
β-Caryophyllene	1411	1.0	32.4	40.5	-	16.1	11.1
Aromadendrene	1431	-	-	-	-	1.2	-
α-Humulene	1445	-	5.1	39.7	-	23.1	24.0
Alloaromadendrene	1451	-	-	-	-	0.5	-
γ-Muurolene	1472	-	-	-	1.8	-	-
(Z,E)-α-Farnesene	1475	-	-	-	21.5	-	-
γ-Selinene	1477	3.8	-	-	-	-	-
Selina-(4,11)-diene	1479	-	-	-	-	0.6	-
β-Selinene	1481	8.8	-	0.4	-	2.9	-
δ-Selinene	1484	-	-	0.5	-	-	-
γ-Amorphene	1487	-	-	-	12.1	-	-
α-Selinene	1490	3.9	-	0.6	-	3.1	-
α-Muurolene	1493	-	-	0.2	-	-	-
β-Bisabolene	1497	-	-	0.7	-	-	-
δ-Amorphene	1504	-	-	-	7.2	-	-
γ-Cadinene	1506	1.3	4.2	0.1	-	-	-
trans-calamenene	1513	-	-	-	-	1.8	-
δ-Cadinene	1515	1.8	10.0	1.4	-	-	0.9
(E)-iso-γ-Bisabolene	1519	-	-	0.1	-	-	-
(Z)-Nerolidol	1523	2.5	-	-	-	-	-
(E)-Nerolidol	1553	-	-	-	6.9	-	-
Isocaryophyllene oxide	1555	1.0	-	-	-	-	-
Caryophyllenyl alcohol	1563	-	-	1.6	2.8	-	-
Spathulenol	1569	-	-	-	-	4.1	-
Caryophyllene oxide	1576	15.4	6.8	1.3	-	7.5	18.9
Globulol	1583	-	-	-	2.1	-	-
Viridiflorol	1585	4.2	-	-	-	0.7	0.8
Salvial-4(14)-en-1-one	1587	3.4	-	-	-	-	-
Widdrol	1589	-	-	1.3	-	-	-
Guaiol	1590	5.0	-	-	-	-	-
1,5,5,8-Tetramethyl, 3,7-cycloundecadien-1-ol	1598	-	-	6.0	-	-	-
Humulene epoxide II	1600	-	-	1.3	-	7.1	17.6
1,10-Di-epi-cubenol	1611	1.5	-	0.9	3.0	-	-
1-Epi-cubenol	1619	-	11.8	-	-	-	-
Epi-α-cadinol	1628	-	-	-	10.2	-	-
Alloaromadendrene epoxide	1629	-	-	-	-	0.4	-
Caryophylla-4(12),8(13)-dien-5-ol	1630	1.1	-	0.2	-	0.4	-
Epi-α-muuralol	1631		4.1	0.5	-		8.2
Selina,3,11-dien-6α-ol	1634	6.7	-	-	-	0.5	-
α-Muurolol	1635	-	-	0.2	-		-
Cubenol	1636	-	-	-	-	0.6	-
δ-Cadinol	1638	-	-	-	-	-	1.8
α-Cadinol	1643	-	-	1.0	4.6	-	12.2
Selina-11-en-4α-ol	1649	13.0	-	-	-	-	-
(*Z*)-14-Hydroxy isocaryophyllene	1655	1.7	-	-	-	-	-
Eudesm-7(11)-en-4-ol	1689	0.8	-	-	-	-	-
Cyclocolorenone	1747	1.9	-	-	-	-	-
Total identified		**82.8**	**80.2**	**98.7**	**72.2**	**91.5**	**97.7**

Monoterpene hydrocarbons	-	-	0.2	-	-	0.4
Oxygenated monoterpenes	-	-	-	-	-	0.5
Total monoterpenes	-	-	0.2	-	-	0.9
Sesquiterpene hydrocarbons	24.6	57.5	84.2	42.6	55.8	37.3
Oxygenated sesquiterpenes	58.2	22.4	14.3	29.6	21.3	59.5
Total sesquiterpenes	82.8	80.2	98.5	72.2	77.1	96.8
Phenyl propanoids	-	-	-	-	14.4	-

RRI: Relative retention index calculated on HP-5 column, with respect to homologous of n-alkanes (C_6-C_{30}, Aldrich Chem. Co. Inc.). *S.arn- Syzygium arnottianum, S.car- Syzygium caryophyllatum, S.hem- Syzygium hemisphericum, S.lat- Syzygium laetum, S.lan- Syzygium lanceolatum, S.zey- Syzygium zeylanicum*

4. Conclusion

Though the genus *Syzygium* includes important spice plants and medicinal plants like *S. aromaticum, S. cumini* and *S. jambos,* most of the Western Ghats endemic *Syzygium* species are least explored for their volatile constituents and bioactivities [13]. Essential oils are important as source of valuable aroma chemicals, flavoring components and bioactive agents and the volatile chemical profiles of *Syzygium* species revealed sesquiterpenoids, particularly caryophyllene isomers and oxygenated derivatives of caryophyllene as the characteristic constituents. Caryophyllene and their derivatives are known for their anti-inflammatory, analgesic, antipyretic, and platelet-inhibitory actions, while its oxide has proven to be cytotoxic [14,15]. The present study is the first report of the leaf volatile constituents of six *Syzygium* species, of which two are endemic to the Western Ghats of south India.

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