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Bud and leaf essential oil composition of *Syzygium aromaticum* from India and Madagascar

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ABSTRACT: The essential oils of *Syzygium aromaticum* were isolated from its buds and leaves by hydrodistillation. The oils were analyzed by high resolution GC and GC-MS. Twenty-eight and 35 constituents representing 99.9% each were identified from the bud oils of Indian and Madagascan origins, respectively. On the other hand leaf oil from Madagascar resulted in the identification of 22 constituents representing 99.9% of the oil. The major constituents in bud and leaf oils were eugenol and β-caryophyllene. Copyright © 2004 John Wiley & Sons, Ltd.

KEY WORDS: *Syzygium aromaticum*; Myrtaceae; bud and leaf oils; essential oil composition; eugenol; β-caryophyllene

Introduction

Syzygium aromaticum (L.) Merril & Perry is an evergreen tree[1] and cloves, clove oil and oleoresin are commercial products. It is native to Molucca Island of Indonesia. The major clove-producing countries are Indonesia, Tanzania, Sri Lanka, Madagascar and, on a limited scale, India. In India it is grown in Kerala, Tamilnadu, Karnataka, Andaman and Nicobar Island over an area of 1735 hectares.[2] The stem, unopened buds and leaves are normally used for extraction of essential oil.[3] Owing to various kind of biological activities, clove oil finds extensive use in dental formulations, tooth paste, breath freshner, mouth washes, soaps, cosmetic items and insect repellent. The oil possess anthelmintic, analgesic, antibacterial, antifungal and anticarcinogenic properties[4–6]. The chief constituent of clove oil is eugenol, which is used as a starting material for the production of vanillin. It has been observed that importance of clove leaf oil is growing due to increased technical use of clove oils. As a part of our studies on the essential oil composition by GC and GC-MS analysis,[7–12] these oils have been examined. A literature survey showed detailed work on chemical composition of bud and leaf essential oils of *S. aromaticum* from different parts of the world,[7,13–30] but there is no significant work on the chemical composition of Madagascar clove leaf oil[25] and Indian clove bud oil,[22] which prompted us to carryout detailed GC and GC-MS analysis of the above oils.

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Experimental

Plant materials

The buds of *Syzygium aromaticum* were procured from a local market, in Lucknow (India), while the buds and leaves oil samples from Madagascar were obtained from M/s Synthite Industrial Chemicals (P) Ltd, Colenchery, Kerala. The buds were hydrodistilled for 4 h in a Clevenger-type apparatus in August 2000. The oil thus obtained was dried over anhydrous sodium sulphate and kept at 4–5 °C in a refrigerator until analysis.

Gas chromatographs (GC)

GC analysis of the oils was performed on a Perkin Elmer GC 8500, using a fused silica capillary column (30 m × 0.32 mm, film thickness 0.25 μm), coated with dimethyl polysiloxane (BP-1). The oven temperature was programmed from 60 to 220 °C at 5 °C/min, then held isothermal at 220 °C for 15 min; injector temperature, 250 °C; detector temperature, 300 °C; carrier gas nitrogen at a inlet pressure of 8 psi; split, 1:80.

Gas chromatography–mass spectrometry (GC-MS)

GC-MS data were obtained on a Shimadzu QP-2000 Mass spectrometer instrument at 70 eV and 250 °C. The GC column was a Ulbon HR-1 (equivalent to OV-1), fused silica capillary column (0.25 mm × 50 m, film thickness 0.25 μm). The initial temperature was 100 °C

for 7 min, and then heated at 5 °C/min to 250 °C. For the Carrier gas helium was used with a flow rate of 2 mL/min.

Identification of compounds

Compounds were identified by comparing the retention indices of the peaks on the BP-1 column with literature values, computer matching against the library spectra build-up using pure substances of components of known essential oils and finally confirmed by comparison of mass spectra with published data.[29–31] The relative amounts of individual components were based on peak areas obtained with FID response factor correction. The retention indices were calculated from gas chromatograms by the logarithmic equation using the homologus series of *n*-alkanes (C-8 to C-22; Poly Science Inc., Niles, USA) as standards.

Results and discussion

The volatile oil was obtained by conventional hydrodistillation of the buds of *S. aromaticum* in a Clevenger type apparatus. The GC and GC/MS analysis resulted in

Table 1. Percentage composition of *Syzygium aromaticum* bud and leaf oils from Madagascar and India

Compound[a]	KI	Bud oil India	Bud oil (Madagascar)	Leaf (Madagascar)
n-Octane	800	—	0.1	—
α-Pinene	932	—	0.1	—
(E)-β-Ocimene	1035	—	t	—
Methyl benzoate	1071	—	t	—
Linalool	1083	0.1	t	—
m-Methyl acetophenone	1132	t	0.1	—
Methyl salicylate	1168	0.3	0.1	0.1
Nerol	1213	t	0.1	—
Carvone	1227	0.1	0.2	0.1
Chavicol	1233	t	0.1	0.1
Linalyl acetate	1243	t	0.1	0.1
Anethole[b]	1259	t	—	—
Eugenol	1340	70.0	82.6	82.0
n-Butyl benzoate	1349	1.3	—	—
α-Cubebene	1359	—	t	—
Methyl eugenol	1364	—	t	—
α-Ylangene	1370	—	t	—
iso-Eugenol-I	1375	0.8	0.1	0.1
Vanillin	1386	t	—	—
α-Copaene	1402	0.1	0.1	—
β-Caryophyllene	1418	19.5	7.2	13.0
(E)-α-Bergamotene	1424	1.3	0.2	0.4
α-Humulene	1450	1.9	0.8	1.5
allo-Aromadendrene	1467	0.3	0.1	—
Germacrene D	1475	0.1	—	—
Eugenyl acetate	1485	2.1	6.0	0.4
α-Selinene	1492	0.1	0.3	—
Calamenene	1508	0.1	0.1	—
γ-Cadinene	1513	0.8	0.2	0.3
δ-Cadinene	1523	0.2	—	—
(E)-Nerolidol	1561	0.1	0.4	0.2
Caryophyllene oxide	1571	0.4	0.3	0.5
Humulene epoxide I	1589	—	0.1	—
Humulene epoxide II	1596	0.1	t	0.1
Cubenol	1606	—	0.1	—
t-Cadinol	1617	0.1	0.1	0.2
t-Muurolol	1634	—	t	—
epi-α-Cadinol	1643	—	0.1	0.1
α-Cadinol	1656	0.1	0.1	0.1
n-Heptadecane	1690	—	0.1	0.2
Benzyl *n*-octanate	1726	—	—	0.1
Myristic acid	1760	—	—	0.1
iso-Propyl myristate	1837	—	—	0.1
Oleic acid	1939	—	—	0.1

[a] Compounds are listed in the order of elution on BP-1 column.
[b] Correct isomer not identified.
t = trace (<0.05%).

the identification of 28 constituents from Indian bud oil, while bud and leaf oils from Madagascar resulted in the identification of 35 and 22 constituents, respectively. The relative concentration of the volatile components identified are presented in Table 1, according to their elution order on the BP-1 column. The major constituents of Indian and Madagascan bud oil were eugenol (70 and 82.6%) and β-caryophyllene (19.5 and 7.2%). From this it is evident that bud oil from Lucknow market was low in eugenol content but higher in caryophyllene content in comparison to Madagascan bud oil. Out of 23 constituents which were common to Indian and Madagascan bud oil, significant differences were observed with respect to eugenyl acetate (2.1 and 6.0%), α-humulene (1.9 and 0.8%), (E) α-bergamotene (1.3 and 0.2%), iso-eugenol-I (0.8 and 0.1%), γ-cadinene (0.8 and 0.2%), (E)-nerolidol (0.1 and 0.4%), allo-aromadendrene (0.3 and 0.1%) and selinene (0.1 and 0.3%).

On comparing our results of bud oil sample procured from Lucknow with those earlier reported from India,[20] variation in the percentage compositions of the main constituents, eugenol (70 and 77.1%) β-caryophyllene (19.5 and 6.2%), eugenyl acetate (2.1 and 5.0%) and α-humulene (1.9 and 1.1%), were observed. The effects of maturity on the composition of bud oil from India[20] revealed that eugenol content increased while eugenyl acetate declined with maturity, probably due to enzymatic hydrolysis of the later into the former. On the other hand our oil matched to a large extent the bud oil of the Malagasy Republic in the percentage composition of eugenol (70 and 72%), β-caryophyllene (19.5 and 15.7%) and α-humulene (1.9 and 1.6%), but differed in the eugenyl acetate (2.1 and 7.8%). Similarly comparison of our Madagascar bud oil results with those earlier reported for Madagascar bud oils[25,20] clearly showed that our oil stands in between those reported by Srinivas[25] and Randriamiharison and Gayadou[26] in the percentage composition of eugenol (82.6, 89.0 and 80.6%), β-caryophyllene (7.2, 4.4 and 10.2%), eugenyl acetate (6.0, 5.5 and 6.6%) and α-humulene (0.8, 0.5 and 1.2%).

Similarly, comparing our results of Madagascan leaf oil with those earlier reported from Madagascar,[25] some variation in the percentage composition of main constituents, eugenol (82.0 and 86.9%), β-caryophyllene (13.0 and 9.9%) and eugenol acetate (0.4 and 1.6%), were observed. Similarly comparison of our results of Madagascan leaf oil with leaf oil of Andaman,[34] India[22] and Indonesia[25] showed similarity to a certain extent in the percentage composition of the main constituents, eugenol (82.0, 81.0, 81.6 and 87.8%), β-caryophyllene (13.0, 12.7, 6.4 and 12.5%), α-humulene (1.5, nil, 1.4 and 1.4%).

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