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Full Length Research Paper

Chemical extracts of leaves and buds of clove (*Syzigium caryophyllatum (L.) Alston*) obtained from trees grown in Bangladesh

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Essential oil obtained by hydrodistillation from fresh leaves and dry buds of *Syzigium caryophyllatum* were analyzed by Gas Chromatography Mass Spectrometry (GC-MS). Thirty eight components were identified in the leaf oil. The main components were eugenol (74.3%), eucalyptol (5.8%), caryophyllene (3.85%) and α-cadinol (2.43%). Thirty one components were identified in bud oil with the main components being eugenol (49.7%), caryophyllene (18.9%), benzene,1-ethyl-3-nitro (11.1%) and benzoic acid,3-(1-methylethyl) (8.9%). The clove oil from Bangladesh was found to be comparable in terms of its eugenol content. It is suggested that clove can be grown as an economically viable crop in Bangladesh.

Key words: *Syzygium caryophyllatum*, bud and leaf oils, essential oil composition, GC-MS, eugenol.

INTRODUCTION

Syzygium caryophyllatum (L.) Alston, (syn. *Syzygium aromaticum* (L.) Merr and Perry commonly called clove, which belongs to the family *Myrtaceae*, is an important aromatic spice. Clove is commercially cultivated in India, Madagascar, Sri Lanka, Indonesia and the south of China. Now-a-days it also cultivated in Bangladesh in a small scale. Clove oil is widely used for flavouring pastry, special sauces and condiments. It is also used in medicines, especially in the preparations for gum and teeth. The tinctures, extracts and oleoresins are also used (Atal and Kapur, 1982). Clove bud oil has biological activities, such as antibacterial, antifungal, insecticidal and antioxidant properties, and are used traditionally as flavouring agent and antimicrobial material in food (Lee and Shibamoto, 2001; Huang et al., 2002; Velluti et al., 2003). The high levels of eugenol contained in clove essential oil responsible for strong antimicrobial activity. This phenolic compound can denature proteins and reacts with cell membrane phospholipids changing their permeability (Briozzo, 1989; Deans and Ritchie, 1987). Clove oil also has several therapeutic effects, including anti-phlogistic, anti-vomiting, analgesic, antispasmodic, anti-carminative, kidney reinforcement, antiseptic and HCMV extracorporeal restraining effect (Liu et al., 1997). In Korea, clove oil is used in aromatherapy and is successfully used for asthma and various allergic disorders by oral administration (Kim et al., 1998).

Clove oil is also widely used as a perfume and food flavouring (Zheng et al., 1992), and as a general antiseptic in medical dental practices (Cai and Wu, 1996). Importantly, Lee and Shibamoto, 2001, reported that clove oil might also be used as an anti-carcinogenic agent due to its antioxidant properties. Their results also suggested that clove oil might be of use as a potential chemopreventative agent. Clove oil is used in the traditional blend of choji (1% clove oil in mineral oil) and is applied to Japanese sword blades to prevent tarnishing of the polished surface (Cai and Wu, 1996; Baytop, 1999). *Syzygium* species have been reported to possess antibacterial (Shafi et al., 2002) and anti-inflammatory activity (Muruganadan et al., 2001). Boulos (1993) reported that the buds of clove were used in folk medicine as diuretic, odontalgic, stomachic, tonicardiac, aromatic condiment properties and condiment with carminative and stimulant activity Several compounds from *S. aromaticum* (namely 5, 7-dihydroxy-2-methylchromone-8-C- -D- glucopyranoside, biflorin, kaempferol, rhamnocitrin, myricetin, gallic acid, ellagic acid and oleanolic acid) have been found to possess growth inhibitory activity against oral pathogens (Cai and

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Wu, 1996). Also, an orsellinic acid glucoside has been isolated from *S. aromaticum* (Charles et al., 1998).

Recently, flavonoid triglycosides have been isolated (Nassar, 2006). The evaluation of antioxidant properties of the raw material allows the determination of its suitability as high quality food beneficial for human health and therefore is of considerable importance. The major constituents in bud and leaf oils were reported to be eugenol and -caryophyllene (Srivastava et al., 2003; Raina et al., 2001; Wenqiang et al., 2007). Kamel et al. (2007) reported the main constituent's flower buds of clove essential oil are phenylpropanoids such as carvacrol, thymol, eugenol and cinnamaldehyde. Amla et al. (2007) also reported that the bud oil contained primarily eugenol, eugenyl acetate and -caryophyllene. This paper reports the results of chemical constituents on clove leaf and bud oils obtained from trees grown in Bangladesh. There are no previous references in literature about these Bangladeshi oils.

MATERIALS AND METHODS

Plant material

The plant materials of clove were collected from the plants grown in the campus of BCSIR Laboratory, Chittagong during June 2009. The specimen was identified by M. Yusuf (Taxonomist) . One-voucher specimen (N-99) was deposited in the herbarium of BCSIR Laboratory, Chittagong.

Extraction of essential oil

Leaves and buds were harvested from healthy, well-grown plants. Freshly_harvested leaves (400 g) and dried buds (300 g) were grounded in a blender separately. The grounded leaves and buds were subjected to hydrodistillation using Clevenger apparatus for 4 h for isolation of oils separately (Clevenger, 1928). The oil samples were stored at 0°C in air- tight containers after drying them over anhydrous sodium sulfate and filtered before going to GC-MS analysis.

GC-MS analysis

The essential oils from leaves and buds of clove were analyzed by GC-MS electron impact ionization (EI) method on GC-17A gas chromatograph (Shimadzu) coupled to a GC-MS QP 5050A Mass Spectrometer (Shimadzu); fused silica capillary column (30 m x 0.25 mm; 0.25 μm film thickness), coated with DB-5 ms (J&W); column temperature 100°C (2 min) to 250°C at the rate of 3°C/min; carrier gas, helium at constant pressure of 90 Kpa. Acquisition parameters full scan; scan range 40 - 350 amu. Samples were injected by splitting and the split ratio 1:20.

Identification of the compounds

Compound identification was done by comparing the NIST library data of the peaks with those reported in literature, mass spectra of the peaks with literature data. Percentage composition was computed from GC peak areas on BD-5 ms column without applying correction factors.

RESULTS

Table 1 shows the relative percentages as single components for clove leaf and bud oil. As shown in the table, 38 and 31 compounds, representing about 96.3 and 98.5% of the essential oil from leaf and bud of clove, were characterized, respectively. The leaf oil contains eugenol (74.28%), eucalyptol (5.78%), caryophyllene (3.85%), α-cadinol (2.43%), limonene (2.08%) and α-caryophyllene (1.52%). The buds oil contains eugenol (49.71%), caryophyllene (18.94%), benzene, 1-ethyl-3-nitro (11.12%), benzoic acid, 3- (1-methylethyl) (8.95%), elixene (3.87%), caryophyllene oxide (1.53%) and -farnesene (1.11%). The identified compounds of the oil and percentage composition are shown in Table 1 where the components are listed in order to their elution on the DB-5 ms column. Results showed that the oils were complex mixture of numerous compounds; many of which were present in trace amounts. It is worth mentioning here that, there is slight variation in the chemical composition of leaf and buds oils.

Eugenol is the main component in both oils. Eugenol, caryophyllene, limonene, eucalyptol, methyl salicylate, chavicol, copaene, alloaromadendrene, germacrene D, α-guaiene and δ-cadinene were observed as the eleven versatile common components present in both the oils with variations in percent content (Table 1). The study reveals that except eugenol as major components, the composition of the oil differs from the earlier reports (Srivastava et al., 2003; Raina et al., 2001; Wenqiang et al., 2007; Kamel et al., 2007) and may, therefore be treated as different chemotypes. On the basis of the above fact, it may be concluded that clove, growing widely in Bangladesh, may be utilized as a source for the isolation of natural eugenol.

The high concentration of eugenol in leaf and buds oil makes it potentially useful in the medicines because they exhibit antibacterial, antifungal, anti-inflammatory activity, insecticidal and antioxidant properties, and are used traditionally as flavouring agent and antimicrobial material in food (Huang et al., 2002; Velluti et al., 2003; Shafi et al., 2002; Muruganadan et al., 2001) . It is worth noting that the clove oil has been reported to be used in folk medicine in the therapeutic effects, including antiphlogistic, antivomiting, analgesic, antispasmodic, anticarminative, kidney reinforcement, antiseptic, HCMV extracorporeal restraining effect, diuretic, odontalgic, stomachic, tonicardiac, aromatic condiment properties and condiment with carminative and stimulant activity.

DISCUSSION

Essential oil from leaf and bud of clove were obtained from hydrodistillation, and their chemical constituents were

Table 1. Chemical constituents of the essential oil from clove leaves and buds.

S/No.	Name of constituents	%	
		Leaf oil	Bud oil
1.	α-Pinene	0.33	--
2.	β-Phellandrene	0.12	--
3.	β-Pinene	0.45	--
4.	α-Phellandrene	0.09	--
5.	α-Terpinene	0.31	--
6.	m-Cymene	0.16	--
7.	Limonene	2.08	0.05
8.	Eucalyptol	5.78	0.06
9.	γ-Terpinene	0.17	--
10	Linalool	0.14	--
11	2-Cyclohexen-1-ol, 1-methyl-4-(1-methylethyl)-	0.04	--
12	2-Heptanol acetate	--	0.05
13	4-Terpineol	0.45	--
14	Methyl Salicylate	0.20	0.25
15	Terpinyl acetate	0.59	--
16	Chavicol	0.08	0.30
17	Eugenol	74.28	49.71
18	Benzyl acetate	--	0.07
19	4-Terpineol	0.45	--
20	Caryophyllene	3.85	18.94
21	Copaene	0.17	0.39
22	α-Caryophyllene	1.52	--
23	Alloaromadendrene	0.05	0.30
24	α-Cubebene	0.02	--
25	Germacrene D	0.38	0.08
26	α-Guaiene	0.06	0.02
27	γ-Elemene	0.21	--
28	β-Bisabolene	0.06	--
29	Benzoic acid, 3-(1-methylethyl)	--	8.95
30	δ-Cadinene	0.21	0.17
31	Benzene, 1-ethyl-3-nitro	--	11.12
32	Guaiene	0.09	--
33	Caryophyllene oxide	0.78	1.53
34	Globulol	0.38	--
35	Ledol	0.16	--
36	Humulane-1,6-dien-3-ol	0.51	--
37	Cedr-9-ene	0.16	--
38	Cubenol	0.19	--
39	Elixene	--	3.87
40	α-Cadinol	2.43	--
41	Megastigma-4,6(E),8(Z)-triene	--	0.05
42	Juniper camphor	0.17	--
43	Kauran-18-al, 17-(acetyloxy)-	0.13	--
44	Alloaromadendrene oxide-(1)	0.11	--
45	-Amorphene	--	0.05
46	Germacrene D	0.38	0.08
47	(+)- Cycloisosativen	--	0.16
48	Nerolidyl acetate	0.06	
49	-Farnesene	--	1.11

Table 1. Contd.

50	-Cadinene	--	0.05
51	Naphthalene,1,2,2,4,4a,7-hexahydro-1,6-dimethyl-4(1-methyethyl	--	0.19
52	Cyclohexane,1,2-dimethyl-3,5-bis(1-methylethenyl)-	--	0.22
53	9 -Acetoxy-3,5 ,8-trimethyltricyclo[6.3.1.0(1,5)]dodec-3-ene	--	0.06
54	Cycloheptane,4-methylene-1-methyl-2-(2-methyl-1-propen-1-yl)-1-vinyl	--	0.08
55	12-Oxabicyclo[9.1.0]dodeca-3,7-diene,1,5,5,8-tetramethyl-,	--	0.11
56	Tetracyclo[6.3.2.0(2,5).0(1,8)]tridecan-9-ol,4,4-dimethyl	--	0.67
57	2',3',4, Trimethoxyacetophenone	--	0.10
58	Benzyl benzoate	--	0.12
59	Squalene	--	0.69

determined by GC-MS. The findings indicated that both the essential oils mainly contain eugenol. The chemical constituents of the Bangladeshi clove leaf and bud oil were found to be comparable to those from clove trees naturally grown in its native regions.

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