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Chemical investigation of the leaf and rhizome essential oils of *Zingiber zerumbet* (L.) Smith from Bangladesh

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Abstract

Zingiber zerumbet (L.) Smith leaf and rhizome oils, obtained by hydrodistillation, were analyzed by gas chromatography mass spectroscopy (GC-MS). Twenty-nine components were identified in the leaf oil. The major components were zerumbone (37.0%); a-caryophyllene (16.4%) and camphene (9.2%). Thirty components were identified in rhizome oil with the main components being in zerumbone (46.8%); a-caryophyllene (19.0%) and 1,5,5,8-tetramethyl-12-oxabicyclo[9.1.0]dodeca-3,7-diene (4.3%). The compositions of both oils varied qualitatively and quantitatively.

Introduction

Zingiber zerumbet (L) Smith, a member of the family Zingiberaceae is well known as *Jangli adha* in Bangladesh. The plant is widely cultivated in village gardens in the tropics for its medicinal properties and as a marketable spice (Saadiah and Halijah, 1995). It grows in the edges of the forests, village thickets in partial shade. It is distributed in Bangladesh, India, Malaysia, Nepal and Srilanka (Anonymous, 1976). It has been reported that plants from this family have anti-inflammatory (Jaganath and Ng, 2000; Somchit and Shukriyah, 2003), anti-ulceration (Mascolo et al., 1989), anti-oxidant (Agrawal et al., 2000) and antimicrobial properties (Nakatani, 2000). It is used to treat stomach aches in Indonesian traditional medicine under the name *Jamu* (Burkill, 1966). In Polynesia it is an ingredient of several medical preparations used to treat ear inflammation and diarrhea (Petard, 1986). It is used in local traditional medicine as a cure for swelling, sores and loss of appetite. The juice of the boiled rhizomes has also been used as a medicine for worm infestation in children. The volatile oil of the rhizomes has been shown to contain zerumbone, humulene and camprene (Hasnah, 1991; Srivastava et al., 2000). On Reunion Island *Z. zerumbet* is grown only in gardens and is used to treat severe sprains in horses and to relieve rheumatic pain. The rhizome oil of *Z. zeruibet* has been the subject of many studies, especially in India.

Dev (1960) isolated and determined the structure of zerumbone. Nigam and Levi (1963) identified, among other constituents, α-humulene, zerumbone, and humulene monoxide. Damodaran and Dev (1968) characterized humulene oxides I, II and III, humulenols I and II, caryophyllene oxide, β-caryophyllene, dihydrophotozerumbone and photo-zerumbone. Chhabra et al. (1975) found zerumbone epoxide. In oil from Fiji, Duve (1980) found highest levels of zerumbone (59%). Dung et al. (1995) found high proportions of (Z)-nerolidol (22-36%), which was absent from rhizomes, in extracts of stems, leaves and flowers, and found zerumbone to predominate in leaves. Srivastava et al. (2000) found in similar proportions curzerenone (14.4%), zerumbone (12.6%) and camphor (12.8%). Chane-Ming et al. (2003) reported that rhizomes were rich in zerumbone (37%), a-humulene (14.4%) and camphene (13.8%) and leaves were rich in trans-nerolidol (21.4%), β-caryophyllene (6.9%) and linalool (7.7%). The characteristics of the oils from the leaves and rhizomes of *Z. zerumbet* allow them to be identified unequivocally. Lechat-Vahirua et al. (1993) reported also the presence of zerumbone (65.3%) as major compounds in the oil from French polynesia. There are no previous references in literature about these Bangladeshi oils. In this work we have determined the chemical composition of leaf and rhizome oils of *Z. zerumbet*. These features allow it to be identified for medicinal use and classified among the other ginger oils available in the international market.

Materials and Methods

Plant material

The plant materials of *Z. zerumbet* were collected from the plants grown in the campus of BCSIR Laboratory, Chittagong during October 2007. A voucher specimen (Y-569) was deposited in the herbarium of BCSIR Laboratory, Chittagong.

Extraction of essential oil

Samples of leaf were harvested from healthy, well-grown, two-year-old plants. Freshly harvested leaves (500 g) and rhizome (200 g) were grounded in a blender separately. The grounded leaves and rhizome were subjected to hydrodistillation using a modified Clevenger-type glass apparatus for 4 hours for isolation of oils separately. 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 oil from leaves and rhizomes of *Z. zerumbet* 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 2.5 mm; 0.25 mm film thickness), coated with DB-5 (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.

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 DB-5 column without applying correction factors.

Results and Discussion

The volatile compounds were identified in the leaves and rhizomes oil of *Z. zerumbet* from Bangladesh were 29 and 30 respectively (Table I). The oil yields were 0.01% and 1.1% respectively. The leaves oil were rich in zerumbone (37.0%); α-caryophyllene (16.4%); camphene (9.2%); 1,2-dihydropyridine,1-(1-oxobutyl),- (5.8%); 3-cyclo hexen-1-carboxaldehyde, 3,4-dimethyl- (3.9%); caryophyllene (3.3%); camphor (2.7%); caryophyllene oxide (2.5%); α-pinene (2.2%); eucalyptol (1.7%) and longipinene, [E]-(1.7%). The rhizome oil was rich in zerumbone (46.8%); α-caryophyllene (19.0%); 1,5,5,8-tetramethyl-12-oxabicyclo[9.1.0]dodeca-3,7-diene (4.3%); caryophy-llene (4.0%); caryophyllene oxide (3.7%); camphene (3.6%); camphor (2.8%); kauran -18-al, 17-(acetyloxy)-, (4.beta.) (2.2%); 1H-cycloprop[e] azulen-4-ol, deca-hydro-1,1,4,7-tetramethyl-, [1ar-(1a.alpha., 4.beta., 4a.beta., 7.alpha.,7a.beta.,7b.alpha.)]- (1.9%); eucalyptol (1.3%) and α-pinene (1.2%). 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 great variation in the chemical composition of leaves and rhizomes oils.

Zerumbone and α-caryophyllene are the main common component in leaves and rhizomes oils. Zerumbone is a anti-cancer bioactive compound (Sharifah et al., 2007). Zerumbone, α-caryophyllene, caryophyllene, camphor, caryophyllene oxide, limonene, α-pinene, eucalyptol, camphene, 3-carene, linalool, borneol, 4-terpineol and cycloheptane, 4-methylene-1-methyl-2-(2-methyl-1-propen-1-1-yl)-1-vinyl were observed as the fourteen versatile common compounds present in both the oils with variations in percent content (Table I). On the other hand, comparison of our oils composition with those reported from different places in the world earlier showed that our oil is especially different to others. But it is very interesting to note that comparison of our results reported on the leaf oil composition from the different places showed different results in the percentage content of some of the major and minor constituents. Zerumbone and α-caryophyllene, which have been reported as major constituents in our oils as well as in almost all the leaves and rhizomes oils of the world (Vahirua et al., 1993; Dung et al., 1993; Chane et al., 2003; Srivastava et al., 2000; Chhabra et al., 1975; Duve, 1980; Nigam and Levi, 1963) were either absent or present in trace amounts in the oil reported. This confirms that the variations in the cultivar reported are not due to geographic divergence and ecological conditions but that is due to different chemotype than ours.

On the basis of above fact it may be concluded that *Z. zerumbet*, growing widely in Bangladesh, may be uti-

Table I

Constituents of leaf and rhizome essential oil from *Zingiber zerumbet*

Peak No.	Name of constituents of leaf essential oil	%	Name of constituents of rhizome essential oil	%
1	Tricyclene	0.6	α-Pinene	1.2
2	α-Pinene	2.2	Camphene	3.6
3	Camphene	9.2	3-Carene	0.8
4	3-Carene	1.0	β-Cymene	0.2
5	Eucalyptol	1.7	Limonene	0.9
6	Limonene	1.1	Eucalyptol	1.3
7	Linalool	0.9	Linalool	0.6
8	Camphor	2.7	Camphor	2.8
9	Borneol	0.5	Borneol	0.3
10	Bornel	0.8	4-Terpineol	0.2
11	4-Terpineol	0.5	β-Terpinyl acetate	0.3
12	α-Terpineol	0.5	Caryophyllene	4.0
13	Caryophyllene	3.3	α-Caryophyllene	19.0
14	α-Caryophyllene	16.4	2,4-Diisopropenyl-1-methylcyclohexane	0.5
15	Cycloheptane, 4-methylene-1-methyl-2-(2-methyl-1-propen-1-1-yl)-1-vinyl	0.7	Anisole, p-styryl-	0.4
16	1,6,10-Dodecatrien-3-ol, 3,7,11-trimethyl, -[S -(z)]	0.5	trans-Nerolidol	0.5
17	Caryophyllene oxide	2.5	Germacrene D-4-ol	0.2
18	1,2-Dihydropyridine,1-(1-oxobutyl),-	5.8	Caryophyllene oxide	3.7
19	3-Cyclohexen-1-carboxaldehyde, 3,4-dimethyl-	3.9	1,5,5,8-Tetramethyl-12-oxabicyclo[9.1.0] dodeca-3,7-diene	4.3
20	Azulene 1,2,3,4,5,6,7,8-octahydro-1,4-dimethyl-7-(1-methylethylidene), -(IS-cis)-	0.5	2-Naphthalenemethanol, 1,2,3,4,4a,5,6,7-octahydro-.alpha.,.alpha.,4a,8-tetramethyl-, (2R-cis)-	0.4
21	2,6-Dimethyl bicyclo [3,2,1]octane	0.7	Bicyclo[3.1.0]hexane-6-methanol, 2-hydroxy-1,4,4-trimethyl-	0.9
22	7-Octylidenebicyclo [4.1.0] heptan-	0.7	Kauran-18-al, 17-(acetyloxy)-, (4.beta.)-	2.2
23	1,5-Cycloundecadien, 8,8-dimethyl-9-methylene	1.1	1H-Cycloprop[e]azulen-4-ol, decahydro-1,1,4,7-tetramethyl-, [1ar-(1a.alpha.,4.beta.,4a.beta., 7.alpha.,7a.beta.,7b.alpha.)]-	1.9
24	3-Isopropyltricyclo [4.3.1.1] (2,5) undec -3-en-l0-ol	0.8	4-Isopropenyl-4,7-dimethyl-1-oxaspiro[2.5] octane	0.2
25	β–Eudesmol	0.7	2-Methylenecholestan-3-ol	1.0
26	Agerospirol	1.0	Carveol	0.9
27	3a,9-Dimethyldodecahydrocyclohepta [d] inden-3-one	0.7	Norethynodrel	0.2
28	trans-Longipinene	1.7	Zerumbone	46.8
29	Zerumbone	37.0	Bicyclo[5.3.0]decane, 2-methylene-5-(1-methylvinyl)-8-methyl-	0.4
30			Cycloheptane, 4-methylene-1-methyl-2-(2-methyl-1-propen-1-yl)-1-vinyl-	0.5

lized as a source for the isolation of natural zerumbone and α-caryophyllene respectively. As a result of this study, the essential oil of *Z. zerumbet* has been extracted and its components identified. The high concentration of zerumbone and α-caryophyllene in leaf and rhizome oil makes it respectively potentially useful in the medicines because they exhibit anti-inflammatory (Jaganath and Ng, 2000; Somchit and Shukriyah, 2003), anti-ulceration (Mascolo et al., 1989), anti-oxidant (Agrawal et al., 2000) and antimicrobial properties (Nakatani, 2000). It is worth noting that the oil of *Z. zerumbet* have been reported to be used in folk medicine in the treatment of inflammation, diarrhea and rheumatic pain.

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