Analysis of the essential oils of the leaves, stems, rhizomes and roots of the medicinal plant *Alpinia galanga* from southern India

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² Department of Chemistry Calicut University, Kerala 673 635, India

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The essential oil of *A. galanga* leaves is rich in 1,8-cineole (28.3%), camphor (15.6%), β -pinene (5.0%), (*E*)-methyl cinnamate (4.6%), bornyl acetate (4.3%) and guaiol (3.5%). The stem essential oil contains 1,8-cineole (31.1%), camphor (11.0%), (*E*)-methyl cinnamate (7.4%), guaiol (4.9%), bornyl acetate (3.6%), β -pinene (3.3%) and α -terpineol (3.3%). 1,8-cineole (28.4%), α -fenchyl acetate (18.4%), camphor (7.7%), (*E*)-methyl cinnamate (4.2%) and guaiol (3.3%) are the main constituents of the rhizome essential oil. The root essential oil contains α -fenchyl acetate (40.9%), 1,8-cineole (9.4%), borneol (6.3%), bornyl acetate (5.4%) and elemol (3.1%).

In addition, biological and aroma effects of the main and minor compounds of the four essential oils of *Alpinia galanga* are discussed in terms of their possible use in medicine, cosmetics and foods.

Keywords: Alpinia galanga (Zingiberaceae), essential oils, leaf, stem, roots, GC, GC-MS

The genus *Alpinia* belongs to the family *Zingiberaceae* with *Alpinia galanga* Willd. syn. *Languas galanga* Stunz (greater galanga) and *Alpinia officinarum* Hance syn. *Languas officinarum* Hance (lesser galangal) as the most important species (1–13). Since *A. officinarum* is mainly known in Asia (China, India, Indonesia, Japan, Malaysia and Thailand), but also in Europe, as a drug prepared from the rhizomes and roots and used as tea or

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tincture with spasmolytic, antiphlogistic and antibacterial effects, also used as stomachicum and against gastric diseases (4, 5, 7), *A. galanga* is an essential spice and food flavoring product as well as a medicament or part of medicaments in Asian folk medicine for various applications, such as against rheumatic oilments, for the treatment of respiratory diseases, as aromaticum and tonicum, but also as aphrodisiacum (2, 3, 8–13).

Intending to analyse the composition and aroma of the essential oils of plants or plant parts from southern India that are used in local foodstuffs or in Indian folk medicine, the aim of this work was to identify the constituents of the essential *A. galanga* oils from the Kerala area to get information about the volatiles responsible for the above mentioned applications. Only few papers report the essential oil components of *Alpinia galanga* samples from *e.g.* India (Bangalore, Hyderabad and Himalaya area), Indonesia, Malaysia and Thailand (2, 3, 8–11, 13). To the best of our knowledge, no information is available about the composition of the essential oil of greater galangal from the Kerala area and its constituents with possible effects useful for food and cosmetic products or medicinal applications.

EXPERIMENTAL

Plant material

The taxon of the plant of *Alpinia galanga* was identified by Dr. A. K. Pradeep (Department of Botany, Calicut University, Kerala, India) and a voucher specimen was deposited in the Herbarium of Calicut University. Fresh leaves, stems, rhizomes and roots of *Alpinia galanga* were collected in the area of the research farm of the Indian Institute of Spice Research in Calicut (July 2000), chopped into small pieces and the essential oils from some hundred grams of leaves, stems, rhizomes and roots were obtained by steam-distillation for 3 hours. The essential oils were dissolved in diethyl ether (*pro analysi*), dried over anhydrous sodium sulphate and the solvent was removed by evaporation (the yields were 0.09% for the leaf oil, 0.1% for the stem oil, 0.23% for the rhizome oil and 0.08% for the root oil).

Olfactoric evaluation

The essential oils were placed on a commercial odor strip (Dragoco Co., Austria) and olfactorically evaluated by professional perfumers.

GC and GC-MS

The GC analyses of the essential oils were performed using a GC-14A with FID connected with a C-R6A-Chromatopac integrator (Shimadzu Co., Japan) and a GC-3700 with FID (Varian Co., Germany) connected with a C-R1B-Chromatopac integrator (Shimadzu). The carrier gas was hydrogen, injector temperature was 250 °C, detector temperature was 320 °C, temperature program 40 °C for 5 min to 280 °C for 5 min with a heating rate of 6 °C min⁻¹. The columns were 30 m × 0.32 mm bonded FSOT-RSL-200 fused silica with the film thickness of 0.25 µm (Biorad Co., Germany) and a 30 m × 0.32 mm bonded Stabilwax with the film thickness of 0.50 µm (Restek Co., USA). Quantifica-

tion was achieved using peak area calculations. Some compounds were identified using retention time correlations with data published elsewhere (14–18).

For GC-MS measurements, a GC-17A with a QP5000 (Shimadzu) and a Compaq-ProLinea data system (class5k-software), a GC-HP5890 with a HP5970-MSD (Hewlett-Packard Co., USA) and ChemStation software on a Pentium PC (Böhm Co., Austria) and a GCQ (Finnigan-Spectronex Co., Germany-USA) with the Gateway-2000-PS75 data system (Siemens-Nixdorf Co., Germany; GCQ-software) were used. The carrier gas was helium, injector temperature was 250 °C, interface heating was 300 °C, ion-source heating 200 °C, EI-mode was 70 eV, scan-range was 41–450 amu with the cycle time of 0.65 s. For other parameters, see the description of GC-FID above. Mass spectra correlations were done using Wiley, NBS, NIST or our own libraries on-line as well as mass spectra databases published elsewhere (15, 19–21).

RESULTS AND DISCUSSION

The odor of the essential oils of *Alpinia galanga* from southern India was described as follows: leaf oil: strong *Eucalyptus*, pinene and camphor notes with herbal side notes; stem oil: strong *Eucalyptus*, camphor and pinene notes with herbal side notes; rhizome oil: strong *Eucalyptus* and camphor notes with intense fresh-spicy-herbal side notes; root oil: strong fresh-spicy with camphoraceous, eucalyptol and borneol notes.

Using gas chromatographic-spectroscopic systems (GC-FID and GC-MS) with different types of columns, more than 115 compounds could be identified, among which mono- and sesquiterpene and cinnamate derivatives were the dominant ones. The main compounds (concentrations higher than 3%, calculated as percent obtained by GC-FID on a FSOT-RSL column) determined in the *A. galanga* leaf oil were 1,8-cineole (28.3%), camphor (15.6%), β-pinene (5.0%), (*E*)-methyl cinnamate (4.6%), bornyl acetate (4.3%) and guaiol (3.5%). The stem oil contained 1,8-cineole (31.1%), camphor (11.0%), (*E*)-methyl cinnamate (7.4%), guaiol (4.9%), bornyl acetate (3.6%), β-pinene (3.3%) and α-terpineol (3.3%) as important constituents, whereas 1,8-cineole (28.4%), α-fenchyl acetate (18.3%), camphor (7.7%), (*E*)-methyl cinnamate (4.2%) and guaiol (3.3%) were found in the rhizome oil. α-Fenchyl acetate (40.9%), 1,8-cineole (9.4%), borneol (6.3%), bornyl acetate (5.4%) and elemol (3.1%) were the main compounds in the root oil (see Table I).

Correlations of the described aroma attributes and thresholds of the identified volatiles (22–26) with the odor impression of the essential *Alpinia galanga* oil point to the following conclusion: the *Eucalyptus* odor notes can be attributed to 1,8-cineole, camphor/camphoraceous/borneol notes are typical of camphor, camphene, borneol and bornyl acetate, (*E*)-methyl cinnamate possesses a herbal note and fresh-spicy notes are characteristic of α -fenchyl acetate, elemol and guaiol. Further minor compounds show only synergistic effects on the described aroma of each essential *A. galanga* oil, but do not develop additional odor directions.

The above composition of the four essential oils of greater galangal from southern India with characteristic odor impressions, suggests the use of these essential *A. galanga* oils in the flavoring of food products, where fresh (direction of *Eucalyptus* and camphor) aromas are required, such as chewing gums and sweets.

Component ^a	KIb	Γc	Sc	RH ^c	ROc
Ethyl acetate	592	nd ^d	0.02	tr ^e	0.01
(E)-2-Hexenal	829	0.12	0.09	nd	nd
(E)-2-Hexenol	852	0.18	0.03	nd	tr
Hexanol	857	0.01	0.14	tr	0.02
Methyl isobutyl ketone	890	tr	nd	nd	nd
α-Thujene	927	0.04	0.02	nd	nd
Tricyclene	929	0.05	tr	0.06	0.04
α-Pinene	937	1.00	0.47	0.55	0.46
Fenchene	943	0.17	tr	0.31	0.05
Camphene	950	2.75	1.61	2.55	2.43
1-Octen-3-ol	964	nd	nd	0.02	0.03
Sabinene	973	0.67	0.03	tr	0.05
β-Pinene	981	4.97	3.29	1.02	0.09
Myrcene	988	0.40	0.58	0.41	1.80
α-Phellandrene	999	0.15	tr	nd	tr
δ-3-Carene	1010	0.41	nd	nd	nd
α-Terpinene	1015	nd	tr	tr	nd
<i>p</i> -Cymene	1019	0.72	0.47	0.61	1.11
Limonene	1025	0.05	0.06	0.14	0.78
1,8-Cineole	1029	28.34	31.12	28.42	9.44
β-Phellandrene	1032	tr	nd	tr	tr
Benzyl alcohol	1034	nd	nd	0.02	0.01
(Z)-β-Ocimene	1037	tr	tr	tr	tr
(E)-β-Ocimene	1039	0.03	0.02	0.02	0.03
γ-Terpinene	1055	0.08	0.05	0.02	0.03
trans-Sabinene hydrate	1060	0.26	0.07	0.04	tr
<i>cis</i> -Linalool oxide	1065	0.17	0.18	0.19	0.11
trans-Linalool oxide	1075	0.09	tr	0.07	0.04
Fenchone	1080	0.07	0.05	1.16	0.53
α -p-Dimethyl styrene	1082	tr	tr	tr	tr
Terpinolene	1084	tr	nd	0.01	0.02
Linalool	1090	0.15	0.25	0.22	0.39
cis-Sabinene hydrate	1092	tr	tr	0.01	nd
α-Fenchol	1100	0.37	0.32	2.21	2.80
Phenylethyl alcohol	1104	tr	tr	nd	nd
β-Fenchol	1108	0.04	0.02	0.13	0.16
Nonanal	1111	nd	nd	0.05	0.01
cis-p-Menth-2-en-1-ol	1113	tr	tr	0.01	0.02
β-Thujone	1115	0.01	0.03	0.01	0.04
Camphor	1125	15.59	11.01	7.71	1.24
trans-Pinocarveol	1131	3.37	2.26	0.98	0.10

 Table I. Essential oils of the leaves (L), stems (S), rhizomes (RH) and roots (RO) of Alpinia galanga from southern India

	Table 1 (continued)							
Component ^a	KIb	Гc	Sc	RH ^c	RO ^c			
β-Terpineol	1135	0.02	0.01	tr	tr			
Isoborneol	1141	0.09	0.15	0.22	0.21			
Isopulegol	1145	tr	0.03	0.08	0.11			
Borneol	1154	1.04	1.07	2.48	6.29			
p-Cymen-8-ol	1167	0.54	0.61	0.52	0.43			
Terpinen-4-ol	1175	1.39	0.66	0.91	1.25			
α-Terpineol	1182	0.74	3.26	2.58	0.77			
Myrtenal	1184	2.18	1.76	0.83	0.12			
Verbenone	1186	0.20	0.43	0.56	0.06			
trans-Carveol	1208	0.46	0.41	0.33	0.26			
α-Fenchyl acetate	1210	0.24	0.26	18.38	40.88			
cis-Carveol	1214	0.22	0.13	0.07	0.22			
Carvone	1228	0.77	0.70	0.89	0.42			
Pulegone	1230	0.03	0.04	0.01	0.06			
Geraniol	1242	0.13	0.06	0.08	0.02			
Linalyl acetate	1249	0.07	0.02	0.03	0.05			
Isopulegyl acetate	1251	tr	tr	nd	0.05			
<i>cis</i> -Sabinyl acetate	1262	0.01	tr	tr	0.05			
2-Hydroxy-1,8-cineole	1267	0.42	1.35	1.31	0.63			
Isobornyl acetate	1274	0.31	0.12	0.09	0.22			
Bornyl acetate	1277	4.28	3.63	1.67	5.35			
Terpinen-4-yl acetate	1281	0.11	0.30	0.11	0.08			
Myrtenol	1283	2.05	1.53	0.67	0.94			
Pinocarvone	1307	0.78	0.82	0.91	0.12			
(Z)-Methyl cinnamate	1320	0.85	0.32	0.13	0.13			
Eugenol	1335	0.19	0.11	0.05	0.01			
(E)-Methyl cinnamate	1352	4.63	7.44	4.22	0.30			
α-Cubebene	1360	tr	0.05	0.04	tr			
α-Copaene	1378	1.07	1.02	0.85	0.74			
β-Patchoulene	1383	0.73	0.66	0.51	0.16			
β-Bourbonene	1386	0.07	0.14	0.33	0.21			
β-Elemene	1399	1.57	1.69	0.71	0.15			
α-Gurjunene	1405	0.48	0.43	0.29	0.24			
β-Caryophyllene	1432	1.52	1.66	0.62	0.83			
β-Gurjunene	1434	tr	0.12	0.04	0.07			
α-Bergamotene	1437	tr	tr	0.09	0.08			
(Z)-β-Farnesene	1448	0.14	0.05	0.09	0.01			
(<i>E</i>)-β-Farnesene	1451	tr	tr	nd	tr			
α-Guaiene	1454	tr	tr	0.06	tr			
Alloaromadendrene	1461	tr	tr	tr	0.08			
α-Humulene	1465	tr	tr	0.07	0.09			
Germacrene D	1469	0.33	0.16	0.16	0.32			

Table I (continued)

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Component ^a	KIb	Lc	Sc	RH ^c	RO ^c			
Ar-Curcumene	1471	tr	nd	nd	nd			
β-Selinene	1477	nd	nd	tr	nd			
Viridiflorene	1481	0.12	0.02	0.08	tr			
γ-Muurolene	1484	0.09	0.01	0.01	0.53			
Valencene	1486	0.01	0.01	0.02	1.07			
α-Muurolene	1488	nd	tr	tr	tr			
α-Selinene	1491	nd	nd	tr	tr			
γ-Elemene	1497	tr	tr	0.02	0.08			
β-Bisabolene	1499	0.03	0.02	0.04	0.03			
γ-Cadinene	1506	0.58	0.74	0.37	0.44			
δ-Cadinene	1515	0.16	0.22	0.18	0.51			
Elemol	1538	1.21	1.94	1.16	3.08			
(E)-Nerolidol	1543	0.24	0.33	0.28	0.42			
β-Chamigrene	1549	0.12	0.30	0.21	0.25			
Carotol	1552	1.17	2.84	1.74	2.64			
Spathulenol	1562	0.30	0.11	0.35	1.07			
Caryophyllene oxide	1578	0.26	0.15	0.05	0.13			
Globulol	1588	0.29	0.24	0.52	0.31			
Ledol	1592	0.06	0.09	0.15	0.43			
Viridiflorol	1596	0.15	0.24	0.36	0.49			
Cubenol	1602	0.36	0.81	0.39	0.67			
Guaiol	1605	3.49	4.87	3.27	1.16			
γ-Eudesmol	1611	0.31	0.14	0.05	0.09			
τ-Cadinol	1624	0.19	0.31	0.18	0.17			
τ-Muurolol	1635	0.07	0.03	0.02	0.15			
α-Cadinol	1641	0.06	0.07	0.14	0.08			
β-Eudesmol	1644	0.55	0.43	0.47	0.59			
α-Bisabolol	1655	0.46	0.54	0.14	0.26			
β-Bisabolol	1667	0.10	0.07	0.01	0.16			
α-Eudesmol	1669	0.22	0.06	0.08	0.20			
(Z)-α-Bergamotol	1692	tr	tr	0.06	tr			
(Z,E)-Farnesol	1697	0.29	0.17	0.07	0.05			
(E,E)-Farnesol	1711	0.11	0.02	tr	0.07			
Nootketone	1802	0.67	1.35	0.85	0.40			

^a Identification on an apolar FSOT-RSL column.

^b Kovats index using an apolar FSOT-RSL column.

^c Concentration calculated as %-peak area obtained by GC-FID analyses.

^d nd – not detected.
 ^e tr – trace compounds (concentration less than 0.01%).

In cosmetics, these aroma systems may be of interest for *e.g.* shower gels, soaps, shampoos and bath products with a fresh top odor.

Medicinal application of essential oils of *Alpinia galanga* from southern India seems to be of special interest in two cases (27–34): (*i*) external treatment of rheumatic pain zones using these oils in creams or pastes will be effective due to the relatively high content of camphor and camphene, which are known for increasing the blood supply of these zones and therefore anti-inflammatory activities, (*ii*) inhalation of the oils when suffering from respiratory diseases will furnish positive effects because of the high content of 1,8-cineole and fenchol derivatives with known antiphlogistic, antibacterial and cooling activities.

CONCLUSIONS

As a result of this comparative investigation of different essential oils (leaves, stems, rhizomes and roots) of *Alpinia galanga* from southern India, it was found that the composition of these oils differed significantly only in quantity, but not in quality. For the first time, more than 115 volatiles of the essential oils from Indian greater galangal were identified. These essential *A. galanga* oils are rich in mono- and sesquiterpenes as well as phenylpropane derivatives. The above mentioned main compounds are also responsible for the characteristic fresh-camphoraceous aroma with herbal and spicy side notes, while the minor components contribute only in a synergistic way. As possible uses of the greater galangal oils from the Kerala area, applications in food flavoring, in cosmetic products (for body care) and in medicaments (treatment of rheumatic diseases and as inhalation agents against respiratory troubles) are proposed.

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$S A \check{Z} E T A K$

Analiza eteričnog ulja iz listova, stabljike, rizoma i korijena biljke Alpinia galanga iz južne Indije

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Istraživano je eterično ulje listova, stabljike, rizoma i korijena biljke *Alpinia galanga* iz južne Indije GC-FID, GC-MS i olfaktometrijom. U sva četiri uzorka identificirani su mono- i seskviterpeni te (*E*)-metil-cinamat. Ti spojevi su odgovorni za karakteristični miris te upotrebu u narodnoj medicini i prehrani.

Eterično ulje listova *A. galanga* sadrži 1,8-cineol (28,3%), kamfor (15,6%), β-pinen (5,0%), (*E*)-metil-cinamat (4,6%), bornil-acetat (4,3%) i gvajol (3,5%). Ulje iz stabljike sadrži 1,8-cineol (31,1%), kamfor (11,0%), (*E*)-metil-cinamat (7,4%), gvajol (4,9%), bornil-acetat (3,6%), β-pinen (3,3%) i α-terpineol (3,3%). 1,8-Cineol (28,4%), α-fencil-acetat (18,4%), kamfor (7.7%), (*E*)-metil-cinamat (4,2%) i gvajol (3,3%) glavni su sastojci ulja iz rizoma. Eterično ulje korijena sastoji se od α-fencil-acetata (40,9%), 1,8-cineola (9,4%), borneola (6,3%), bornil-acetata (5,4%) i elemola (3,1%). U radu se diskutira o mogućoj primjeni eteričnog ulja biljke *Alpinia galanga* u medicini, kozmetici i prehrani.

Ključne riječi: Alpinia galanga (Zingiberaceae), eterična ulja, list, stabljika, korijen, GC, GC-MS

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