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Chemical Composition of *Cinnamomum* Species Collected in Sarawak

(Komposisi Kimia *Cinnamomum* Spesies dari Sarawak)

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ABSTRACT

Cinnamomum *species (Lauraceae) are well known for their fragrance and medicinal value. The essential oils of three* Cinnamomum *species (*C. macrophyllum, C. crassinervium *and* C. griffithii*) collected in Sarawak were obtained by hydrodistillation and analyzed by gas chromatograpy mass spectrometry (GC-MS). The analysis of the oils showed that most of the essential oils were mainly phenylpropanoids and monoterpenes with a small amount of sesquiterpenes present. Both* C. griffithii *and* C. crassinervium *contained similar major chemical composition such as* β*-linalool, methyl cinnamate and eugenol methyl ether. No presence of methyl cinnamate and* β*-linalool were found in the oil of* C. macrophyllum. m*-Eugenol was prominent in the leaf oil of* C. macrophyllum*, while cinnamaldehyde was found mainly in the bark oil of* C. macrophyllum. *High percentage of camphor was identified in the bark and root oil of* C. macrophyllum*, compared to small amount of camphor found in the both root oil of* C. griffithii *and* C. crassinervium.

Keywords: Cinnamomum *species; essential oil; monoterpene; phenylpropanoid*

ABSTRAK

Spesies tumbuhan Cinnamomum *(Lauraceae) terkenal dengan aroma dan nilai perubatan. Minyak pati daripada tiga spesies* Cinnamomum (C. macrophyllum, C. crassinervium *dan* C. griffithii*) telah diperoleh melalui penyulingan hidro dan dianalisis oleh kromatografi gas spektrometri jisim (GC-MS). Analisis minyak menunjukkan bahawa sebahagian besar minyak pati kebanyakannya daripada fenilpropanoid dan monoterpena dengan sedikit kehadiran siskuiterpena. Kedua-dua spesies* C. griffithii *dan* C. crassinervium *mengandungi komposisi kimia utama yang sama seperti β-linalool, metil simanat dan eugenol metil eter. Tiada kehadiran metil simanat dan β-linalool ditemui dalam minyak C.* macrophyllum. m*-eugenol didapati dengan banyak di dalam minyak daun* C. macrophyllum*, manakala sinnamaldihid dikenal pasti terutamanya di dalam minyak kulit* C. macrophyllum. *Peratusan kamfor yang tinggi telah dikenal pasti dalam kulit dan akar minyak* C. macrophyllum*, berbanding dengan sedikit kamfor yang terdapat di dalam minyak akar kedua-dua* C. griffithii *dan* C. crassinervium.

Kata kunci: Fenilpropanoid; minyak pati; monoterpena; spesies Cinnamomum

INTRODUCTION

Cinnamomum belongs to the Lauraceae family and have been studied extensively for their essential oil constituents. The genus *Cinnamomum* comprises approximately 250 species in the tropical and subtropical regions, mostly in Asia and some in South and Central America, and Australia (Mabberley 2008). *Cinnamomum* has trinerved and fragrant leaves, fruits seated on a cupule and paniculate inflorescences, flower with nine stamens (Soh 2011). Twenty-one species are found in Peninsular Malaysia (Ibrahim et al. 1995) while thirty-six species are found in Borneo (Soh 2011). Some of the species that are found in Borneo are *C. corneri*, *C. crassinervium*, *C. calciphilum*, *C. pendulum*, *C. politum*, *C. suavenium*, *C. subcuneatum*, *C. tahijanum, C. verum*, *C. burmannii*, *C. grandifolium*, *C. kinabaluense* and *C. sintoc* (Soh 2011). *Cinnamomum* has been used as spice and food flavoring around the world and the usage is not only for the flavors, but also for its health benefits. Research interest has focused on the essential oil of the species with chemopreventive, antibacterial, hypolipidemic and antiplatelet properties (Craig 1999).

Several essential oils of *Cinnamomum* species have been investigated for their chemical components. For example, stem bark oil of *C. tahijanum* and *C. iners* were found rich in eucalyptol (Baruah et al. 2001; Nor Azah et al. 1999). While the leaf oil of *C. tamala* and *C. zeylanicum* and bark oil of *C. sintoc*, were prominent with eugenol (Fischer 1960; Ibrahim et al. 1994; Mallavarapu et al. 1995). Cinnamaldehyde, a yellowish strong odour oil was found dominant in the bark oil of *C. tamala*, stem-bark oil *C. zeylanicum,* leaf oil of *C. cassia* and leaf oil of *C. burmanii* (Fischer 1960; Lee et al. 2004; Senanayake et al. 1978; Wang et al. 2008). The compound has been widely used in medicine, food, cosmetic and anti-fungal agent to prevent food spoilage (Bown 1995; Fabio et al. 2003). Camphor was found abundantly in the root bark oil of *C. zeylanicum* and leaf oil of *C. longepetiolatum* (Senanayake et al. 1978; Tran et al. 2008). Camphor is used topically to reduce pain and treat fungal infections.

Although there are several reports on *Cinnamomum* sp. oils, there are no information available for *C. macrophyllum, C. crassinervium* and *C. griffithii*. In this study, chemical composition of the essential oils of *C. macrophyllum, C. crassinervium* and *C. griffithii* growing in Sarawak, Malaysia was investigated.

Materials and Methods

Plant material and oil isolation

The fresh *C. crassinervium* and *C. griffithii* were collected from Bau while *C. macrophyllum* was collected from Sematan, Sarawak for taxonomic identification and laboratory analyses. Voucher specimens for each plant (*C. macrophyllum* FBAUMS-86, *C. crassinervium* FBAUMS 49 and *C. griffithii* FBAUMS-65) were deposited at the Herbarium Department of UNIMAS. The plants were separated into fruit, leaf, bark and root. 150 g of each fresh sample was subjected to hydrodistillation in a Clevenger-type apparatus for 7 h. The essential oils obtained were collected and dried over anhydrous sodium sulphate to make moisture free and refrigerated in the dark at 4°C.

Analysis of the oils

Qualitative and quantitative analysis of each essential oil sample was performed on Shimadzu QP-5000 GC-MS using medium polarity capillary column BPX-5 (30 cm length × diameter 0.25 mm, film thickness of 0.25 μm). Helium gas was used as the carrier gas. The initial temperature was programmed at 50ºC and increased to 280ºC with the rate of 6ºC/min and held for 10 min at the final temperature. The temperature for the injector and detector was set at 280ºC and 300ºC, respectively. The identification of compounds was based on a comparison of their retention indices and mass spectra with those found in the literature (Arce & Arn 2010) and supplemented by data in the National Institute of Standards and Technology (NIST). Homologous series of n-alkanes were used as standards. The relative proportions of the essential oil constituents were expressed as percentages obtained by peak area normalization measurements.

Results and Discussion

The hydrodistillation of *Cinnamomum* species gave brown and yellowish oil yield ranging from 0.27% - 3.60% (v/w), on dry weight basis, for different plant parts (Table 1). The essential oil from leaf of *C. griffithii* gave the highest yield among all plant parts, which was 3.60%. Root of *C. macrophyllum* contributed about 2.76% while 2.67% and 1.80% of the total oil yield could be recovered, respectively, from leaf of *C. crassinervium* and bark of *C. griffithii*. The lowest oil yield was obtained from root of *C. crassinervium* (0.27%).

Table 2 shows the chemical compositions identified in the essential oils isolated from various parts of *C. macrophyllum, C. crassinervium* and *C. griffithii*. Among the species, the essential oil of *C. macrophyllum* gave the largest number of identified constituents except for the leaf oil. The fruit oil of *C. macrophyllum* gave 21 compounds, representing 91% of the total oil composition and was rich in sesquiterpene. The most significant compounds in the fruit oil were α-cadinol (19.70%) and epi-bicyclosesquiphellandrene (12.11%). 3% of the oil was made of monoterpenes. The bark oil of *C. macrophyllum* was mainly composed of phenylpropanoids (50.39%) and monoterpenes (27.16%) with a small percentage of sesquiterpenes (8.59%). The major constituents were trans-cinnamaldehyde (32.12%), camphor (14.35%) and safrole (17.58%). The leaf oil of *C. macrophyllum* gave only 4 identified compounds, representing 100% of total oil. The oil was dominated mainly by phenylpropanoids, which were *m*-eugenol (91.84%) and trans-cinnamaldehyde (1.93%), with a low percentage of sesquiterpenes (caryophyllene, 3.34% and caryophyllene oxide, 2.89%). The root oil of *C. macrophyllum* was rich in camphor (38.55%), followed by d-limonene (13.08%), eucalyptol (11.91%) and β-myrcene (9.20%). Only two sesquiterpenes

TABLE 1. Oil yield obtained from *Cinnamomum* species by using hydrodistilation

Species	Part	Colour	Oil yield (%)
C. macrophyllum	Leaf	Yellow	0.90±0.01
	Fruit	Yellow	0.58
	Bark	Brown	1.25±0.33
	Root	Yellow	2.76±0.22
C. griffithii	Leaf	Yellow	3.60±0.21
	Stem	Yellow	0.47
	Bark	Brown	1.80±0.30
	Root	Yellow	0.50
C. crassinervium	Leaf	Yellow	2.67±0.18
	Stem	Yellow	0.47
	Bark	Brown	0.40±0.03
	Root	Yellow	0.27

*Percentage oil yield (v/w) was calculated as oil volume (mL)/ dry plant samples (g) × 100

TABLE 2. Phytochemical compositions of *Cinnamomum* sp. essential oil from different parts of plant

Chemical compound	RI[a]	*C. macrophyllum*				*C. griffithii*				*C. crassinervium*				Method of identification[b]
		Bark	Leaf	Root	Fruit	Bark	Leaf	Stem	Root	Bark	Leaf	Stem	Root	
Monoterpene														
β-Myrcene	993			9.20										RI, MS
α-Phellandrene	1008			3.14										RI, MS
β-Linalool	1110					23.51	84.49	22.69	1.24	5.93	94.57	5.32		RI, MS
α-Terpinene	1022	1.62		3.20										RI, MS
o-Cymol	1030			1.82										RI, MS
d-Limonene	1035	1.27		13.08										RI, MS
Eucalyptol	1037			11.91					0.48					RI, MS
β-Phellandrene	1038	5.24												RI, MS
β-Trans-Ocimene	1040			2.01										RI, MS
γ-Terpinene	1064			2.57										RI, MS
Benzylacetaldehyde	1066	1.43												RI, MS
4-Carene	1090			2.46										RI, MS
Fenchyl alcohol	1138				0.59									RI, MS
Borneol	1157				0.58									RI, MS
α-Terpineol	1210	1.29		3.02	1.83		1.74	1.30	1.52					RI, MS
Camphor	1213	14.35		38.55					0.32				2.48	RI, MS
Terpineol-4-ol	1214	1.96		2.98										RI, MS
Phenylpropanoid														
trans-Cinnamaldehyde	1248	32.12	1.93											RI, MS
Safrole	1270	17.58						1.25	29.36				16.80	RI, MS
Methyl cinnamate	1375					52.68		48.13	3.10	21.07		60.62		RI, MS
m-Eugenol	1392	0.69	91.84											RI, MS
Methyl eugenol	1423					14.09		17.31	59.01	57.49		34.06	74.94	RI, MS

(continue)

Continued (TABLE 2)

Chemical compound	RI[a]	*C. macrophyllum*				*C. griffithii*				*C. crassinervium*				Method of identification[b]
		Bark	Leaf	Root	Fruit	Bark	Leaf	Stem	Root	Bark	Leaf	Stem	Root	
Sesquiterpene														
α-Cedrene	1371				3.53									RI, MS
Copaene	1387				1.43									RI, MS
Aristolene	1427			4.90										RI, MS
Caryophyllene	1433		3.34	0.86	4.30	1.38		0.96			2.29			RI, MS
α-Bergamotene	1440					0.92								RI, MS
Cinnamyl acetate	1457	0.91												RI, MS
Aromadendrene	1477				2.28									RI, MS
Epi-bicyclosesquiphellandrene	1485				12.11									RI, MS
γ-Muurolene	1489				1.93									RI, MS
α-Cubebene	1494	2.50												RI, MS
α-Muurolene	1514				9.54									RI, MS
Bisabolene	1516					1.88								RI, MS
Ledene	1507				4.01									RI, MS
γ-Cadinene	1531	4.10			8.86									RI, MS
Calamenene	1540				3.22									RI, MS
Cadala-1(10),3,8-triene	1562	1.08			0.65									RI, MS
Elemicin	1563					2.45		4.18	3.34	8.39				RI, MS
trans-Nerolidol	1569				0.91		2.54	1.04						RI, MS
Cubenol	1579				3.06									RI, MS
Caryophyllene oxide	1578		2.89		6.67									RI, MS
Globulol	1581				4.10									RI, MS
Guaiol	1600				0.63									RI, MS
Agarospirol	1629				1.07									RI, MS
α-Cadinol	1650				19.70									RI, MS
Total identified		86.14	100.00	99.70	91.00	96.91	88.77	96.86	98.37	92.88	96.86	100.00	94.22	

RI[a]: retention indices on BPX-5 column

[b]The components of the essential oil were identified by comparisons of their mass spectra with those in a computer library (MS) or confirmed by comparisons of their retention indices (RI) with data published in a reference book (Arce & Arn 2010).

were detected in the root oil which were aristolene (4.90%) and caryophyllene (0.86%).

The essential oils of *C. crassinervium* and *C. griffithii* gave less than eight compounds, representing 88% to 100% of the total oil composition. Most of the *C. crassinervium* and *C. griffithii* oils were mainly dominated by monoterpenes and phenylpropanoids. The bark oil of *C. griffithii* was composed of methyl cinnamate (52.68%), β-linalool (23.51%) and methyl eugenol (14.09%). Low percentage of sesquiterpenes (6.63%) was found in the bark oil. High percentage of methyl cinnamate (48.13%) was presence in the stem oil. Other compounds present in the stem oil were β-linalool (22.69%) and methyl eugenol (17.31%). The leaf oil was dominated with β-linalool (84.49%), however no phenylpropanoid was found in the oil. Other compounds that were presence in the leaf oil were α-terpineol (1.74%) and trans-nerolidol (2.54%). The root oil was rich in methyl eugenol (59.01%) and safrole (29.36%) with low amount of methyl cinnamate (3.10%) and β-linalool (1.24%). Methyl eugenol was the major compound in the bark, stem and root oil of *C. crassinervium*. The leaf oil was rich in β-linalool (94.57%) and caryophyllene (2.29%). Bark and stem oil of *C. crassinervium* were mainly composed of methyl cinnamate and methyl eugenol, with low amount of β-linalool. In the bark oil, methyl eugenol (57.49%) was higher compared to methyl cinnamate (21.07%), while the stem oil has higher constituent of methyl cinnamate (60.62%) compared to methyl eugenol (34.06%). Root oil was dominated by methyl eugenol (74.94%) and safrole (16.80%). Low percentage of elemicin was detected in bark (8.39%) and root oil (2.01%).

No previous study on essential oil of *C. macrophyllum*, *C. griffithii* and *C. crassinervium* has been reported. However according to Ragasa et al. (2013), dichloromethane extract of leaf of *C. griffithii* showed the presence of benzyl benzoate as main composition. In this study, high percentage of methyl cinnamate was identified in the bark oil of *C. griffithii,* which was similar to the chemical composition of bark oil of *Cinnamomum rhyncophyllum* (Ibrahim et al. 2008). Methyl cinnamate was also identified as major constituent in the bark oil of *C. impressicostatum* and *C. pubescens* (Ali et al. 2010). The high percentage of linalool in the leaf oil of *C. griffithii* and *C. crassinervium* were similar to the previous study on the leaf oil of *C. camphora,* which showed linalool (95%) as major chemical composition with other constituents representing less than 1% (Caren et al. 1999). The result was also significant as described by Lin et al. (1987) and Tao et al. (1987), where linalool was the main constituent in the leaf oil of *C. camphora* with percentage varied from 66-91%.

High percentage of cinnamaldehyde (32.12%) and camphor (14.35%) were detected in the bark oil of *C. macrophyllum* in this study. Similarly, cinnamaldehyde, was found prominent in the bark oil of *C. tamala* and stem-bark oil *C. zeylanicum,* (Fischer 1960; Senanayake et al. 1978). Apart from that, the current study showed the leaf and root oil of *C. macrophyllum* was dominated with eugenol and camphor, respectively. And the finding was similar to the leaf and root oil of *C. zeylanicum* (Paranagama et al. 2001).

Based on the result obtained, both of the leaf oil of *C. griffithii* and *C. crassinervium* could be a good source of β-linalool. Linalool is a potential agent for anti-inflammation and anticancer activity (Chang & Shen 2014; Peana et al. 2001). The essential oils of leaf of *C. macrophyllum,* bark of *C. crassinervium* and root of both *C. griffithii* and *C. crassinervium* could also be studied as an alternative drug for treating cardiovascular diseases due to the presence of eugenol. Apart of having antimicrobial activity, according to Naderi et al. (2004), eugenol gave the highest antioxidative activity against LDL oxidation and can change the affinity of the LDL particles for the LDL receptor.

Conclusion

The three *Cinnamomum* species in this study, showed that most of the essential oils were mainly made of phenylpropanoids and monoterpenes with small amounts of sesquiterpenes present. Overall, the results obtained could contribute to the knowledge of the chemical composition of the *Cinnamomum* species collected in Sarawak. The oils can be of interest for further study of their bioactivity as alternative drugs in healthcare.

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