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Volatile Constituents from *Cinnamomum zeylanicum* Fruit Stalks and Their Antioxidant Activities

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Cinnamomum zeylanicum Blume is an important spice and aromatic crop having wide applications in flavoring, perfumery, beverages, and medicines. The steam-distilled volatile oil from cinnamon fruit stalks was analyzed with GC and GC-MS. It showed the presence of hydrocarbons (44.7%) and oxygenated compounds (52.6%). Twenty-seven compounds constituting ca. 95.98% of the volatile oil were characterized. (*E*)-Cinnamyl acetate (36.59%) and (*E*)-caryophyllene (22.36%) are found to be major compounds. The volatile oil was screened for its potential as an antioxidant by using in vitro models, such as the β-carotene-linoleate and phosphomolybdenum complex method. The volatile oil showed 55.94% and 66.9% antioxidant activity at 100 and 200 ppm concentration, respectively. Also, the volatile oil showed good antioxidant capacity, using the formation of the phosphomolybdenum complex. A comparison of the chemical composition of the volatile oil was made with that of buds, flowers, and fruits. This is the first report on the chemical composition of volatile oil of the fruit stalks of this species and its antioxidant activity.

KEYWORDS: ***Cinnamomum zeylanicum*; (*E*)-cinnamyl acetate; (*E*)-caryophyllene; volatile oil; β-carotene-linoleate; phosphomolybdenum; antioxidant activity; GC-MS**

INTRODUCTION

Lauraceae is an economically important family consisting mostly of trees or tree-like shrubs. The genus *Cinnamomum* comprises about 250 species which are distributed in Asia and Australia. The trees occur in South India up to altitudes of 500 m, but mostly below 200 m. The trees flower in January and fruits ripen during May−August (*1*). *Cinnamomum zeylanicum* (*C. zeylanicum*), the source of cinnamon bark, leaf, and their essential oils, is an indigenous tree of Sri Lanka. Many species of cinnamon yield a volatile oil on distillation. The most important cinnamon oils in world trade are those from *C. zeylanicum*, *C. cassia*, and *C. camphora*. The other species provide oils which are utilized as sources for chemical isolates. However, a number of other cinnamon species are distilled on a much smaller scale and the oils used either locally or exported (*1*). Cinnamon leaf and bark are used as spices and in the production of essential oils. Leaves have a hot taste and emit a spicy odor when crushed. Cinnamon offers a variety of oils with different aroma characteristics and composition to the flavor industry. The root bark was reported to have camphor as the main constituents, but does not seem to have commercial value, unlike leaf and stem bark oils (*2*). Cinnamon leaf oil has a warm, spicy, but rather harsh odor, lacking the rich body of the bark oil. Leaf oil has a fragrant odor and a very pungent taste.

Mallavarapu et al. (*3*) identified 53 constituents along with eugenol (81−84.5%) as major components in cinnamon leaf oil. The fruits of the cinnamon are also aromatic and possess a sweet spicy aroma. Thirty-four compounds have been identified previously in cinnamon fruit oil with (*E*)-cinnamyl acetate (42−54%) and (*E*)-caryophyllene (9−14%) as the major components (*4*). Möllenbeck et al. (*5*) reported the Madagascar origin cinnamon essential oil composition and enantiomers using capillary gas chromatography coupled to mass spectrometry (HRGC-MS). It was found that 1,8-cineole (62.4%) and *trans*-cinnamaldehyde (41.3%) are the major compounds in *C. camphora* and *C. zeylanicum*, respectively. Furthermore, by using chiral GC the enantiomeric distributions of linalool and terpinen-4-ol were determined to be (3*R*)-(−)-linalool (95%), (3*S*)-(+)-linalool (5%) and (4*R*)-(−)-terpinen-4-ol (69%), (4*S*)-(+)-terpinen-4-ol (31%), respectively. Baratta et al. (*6*) reported the antimicrobial and antioxidant properties of commercial cinnamon oil. It was found that *trans*-cinnamaldehyde was the major compound and the cinnamon oil was found to be most active in inhibiting the growth of all the tested bacterial strains. Twenty-six compounds constitute 97% of the volatile oil from cinnamon flowers and it was found to contain (*E*)-cinnamyl acetate (42%), (*E*)-α-bergamotene (8%), and caryophyllene oxide (7%) as the principal compounds (*7*). Raina et al. (*8*) reported the 47 chemical constituents of *C. zeylanicum* leaves from little Andaman (India) with eugenol (76.6%) as the major compound. Recently, 34 components representing 98% of the

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oil were characterized in cinnamon buds with α-copane (23%) and α-bergamotene (27.4%) being the major constituents (*9*).

Currently synthetic antioxidants have been suspected to cause or promote negative health effects, hence stronger restrictions have been placed on their application and there is a trend to substitute them with naturally occurring antioxidants (*10*). Some natural antioxidants (e.g. rosemary and sage) are already exploited commercially either as antioxidant additives or as nutritional supplements (*11*).

A literature survey revealed no reports on the chemical composition of volatile oil from cinnamon fruit stalks. Hence, we report the results of the GC-MS analysis of the volatile oil from cinnamon fruit stalks and its antioxidant activity for the first time.

EXPERIMENTAL PROCEDURES

Materials. *trans*-Cinnamaldehyde, *trans*-cinnamyl acetate, and benzyl benzoate were obtained from M/s Sigma-Aldrich Corporation (Bangalore, India). The fruit stalks of *Cinnamomum zeylanicum* were collected from Karkala (South Kanara district, Karnataka State, India) during the fruit-ripening stage. The fruit stalks were dark purple and 1.1 to 4.0 cm long. The species was identified and a voucher specimen was deposited at the Manasagangotri herbarium (MGH NO.4A/96/02), Department of Botany, University of Mysore, Mysore.

Isolation of Volatile Oil. Cinnamon fruit stalks (50 g) were coarsely powdered in a mixer grinder, 150 mL of ice-cold water was added, and the mixer grinder was run for 2 min before the mixture was transferred to a 500-mL round-bottom flask. Then the Clevenger apparatus along with a condenser was assembled and the mixture was subjected to hydro-distillation for 4 h. The yield of volatile oil was 0.40 mL. The oil was dried over anhydrous sodium sulfate and stored at 4 °C until analyzed. The light yellow oil possessed a sweet floral odor.

Purification of (*E*)-Cinnamyl Acetate. One milliliter of fruit stalk volatile oil was impregnated on 2 g of silica gel and loaded on a 20-g silica gel column for chromatography. The column was eluted with 100 mL of nonpolar solvent. Then the polarity was increased with medium polar halogentated solvent (100 mL). The eluates were concentrated under reduced pressure at 30 °C to obtain (*E*)-cinnamyl acetate at a yield of 0.31 mL (*12*).

Gas Chromatography Analysis. The volatile oil was analyzed on a Shimadzu GC-15A (Kyoto, Japan) chromatograph equipped with a FID detector and SE-30 column (3.0 m × 3 mm), made up of 3% SE-30 on 80/10 superlcoport packing, methyl silicone phase type. The oven temperature was programmed from 75 °C for 5 min to 225 °C at 3 °C/min and held for 3 min at maximum temperature; the injector port temperature was 225 °C; the detector temperature was 250 °C; and nitrogen was used as the carrier gas at 40 mL/min. Peak areas were computed by a Shimadzu Chromatopac integrator.

Gas Chromatography−Mass Spectrometry (GC-MS) Analysis. The volatile oil sample was analyzed on a Shimadzu GC-17A (Kyoto, Japan) chromatograph equipped with a QP-5000 (Quadrapole) mass spectrometer. The oil sample was diluted 25 times with acetone (1 μL) was injected. A fused silica capillary column SPB^{TM-1} (30 m × 0.32 mm i.d., film thickness 0.25 μm) coated with poly(dimethylsiloxane) was used. Helium was used as the carrier gas at a flow rate of 1 mL/min; the injector port temperature was 225 °C; the detector temperature was 250 °C; the oven temperature was kept initially at 60 °C for 2 min and then increased to 250 °C at the rate of 2 °C/min, at which temperature the column was maintained for 5 min; the split ratio was 1:50. Mass spectra were recorded under electron impact ionization at 70 eV electron energy with a mass range from 40 to 400 at a rate of one scan/second. Essential oil constituents were identified by comparing retention times of the GC peaks with those of reference compounds run under identical conditions and by comparison of retention indices with literature data (*13, 14*) and by matching their fragmentation pattern in mass spectra with those of NIST62-LIB library and published mass spectra (*15, 16*).

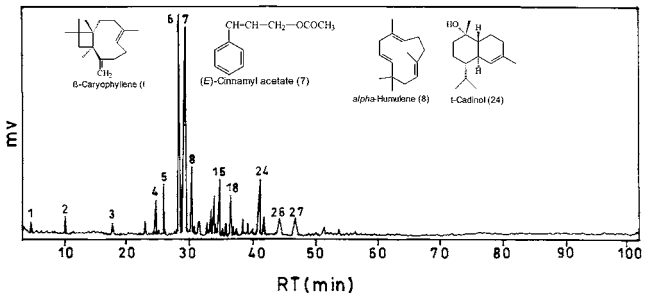

Figure 1. GC-MS total ion chromatogram of cinnamon fruit stalk volatile oil and structures of four major compounds. The peak numbers correspond to the numbers in **Table 1**.

NMR Analysis. ^{1}H NMR (90 MHz, $CDCl_3$) spectra were recorded on a Varian EM-390 instrument. Tetramethylsilane (TMS) was used as the internal standard.

Antioxidant Assay with the β-Carotene-linoleate Model System. The antioxidant activity of fruit stalk volatile oil and propyl gallate (PG) was evaluated by the method of Jayaprakasha et al. (*17*). A 0.2-mg sample of β-carotene in 0.2 mL of chloroform, 20 mg of linoleic acid, and 200 mg of Tween-40 (polyoxyethylene sorbitan monopalmitate) were mixed. Chloroform was removed at 40 °C under vacuum, and the resulting mixture was diluted with 10 mL of water and mixed well. To this emulsion was added 40 mL of oxygenated water. Four-milliliter aliquots of the emulsion were pipetted into different test tubes containing 0.2 mL of diluted volatile oil (equivalent to 100 and 200 ppm) and propyl gallate (equivalent to 100 and 200 ppm) in ethanol. PG was used for comparative purposes. A control containing 0.2 mL of ethanol and 4 mL of the above emulsion was prepared. The tubes were placed at 50 °C in a water bath and the absorbance at 470 nm was taken at zero time ($t = 0$). The measurement of absorbance was continued at intervals of 30 min until the color of β-carotene disappeared in the control tubes ($t = 210$ min). A mixture prepared as above without β-carotene served as the blank. All determinations were carried out in triplicate. The antioxidant activity (AA) of the extracts was evaluated in terms of bleaching of the β-carotene, using the following formula, $AA = 100[1 - (A_o - A_t)/(A°_o - A°_t)]$, where A_o and $A°_o$ are the absorbance values measured at zero time of the incubation for the test sample and the control, respectively. A_t and $A°_t$ are the absorbance values measured in the test sample and the control, respectively, after incubation for 210 min.

Antioxidant Capacity by the Phosphomolybdenum Method. The total antioxidant capacity of fruit stalk volatile oil and PG was evaluated by the method of Prieto et al. (*18*). An aliquot of 0.1 mL of sample solution (equivalent to 100 and 200 ppm) was combined with 1 mL of reagent solution (0.6 M sulfuric acid, 28 mM sodium phosphate, and 4 mM ammonium molybdate). In the case of the blank, 0.1 mL of methanol was used in place of sample. The tubes were capped and incubated in a boiling water bath at 95 °C for 90 min. After the samples had cooled to room temperature, the absorbance of the aqueous solution of each was measured at 695 nm against a blank in a Genesys-5-UV−visible spectrophotometer (Milton Roy, New York). For samples of unknown composition, fat-soluble antioxidant capacity was expressed as equivalents of α-tocopherol (mmol/g of oil) (*18*).

RESULTS AND DISCUSSION

The hydro-distilled cinnamon fruit stalk volatile oil was analyzed with GC and GC-MS. The GC-MS total ion chromatogram and four major compounds from the volatile oil are presented in **Figure 1**. In the volatile oil of fruit stalks of *C. zeylanicum* 27 constituents were identified (**Table 1**). These compounds constituted more than 95% of the volatile oil. Retention indices for all the compounds were determined according to the Kovats method, using *n*-alkanes as standards (*13*).

Oxygenated compounds are present to the extent of 52.6%. Three types of oxygenated compounds are present, viz., esters

Table 1. Chemical Composition of *Cinnamomum zeylanicum* Fruit Stalk Volatile Oil by GC-MS

peak no.	RT (min)	compd	peak area (%)	KI[a]	identification method[b]
1	4.40	β-pinene	0.40	950	RI, MS
2	10.07	linalool	0.70	1051	RI, MS
3	17.53	(*E*)-cinnamaldehyde	0.34	1240	RI, MS, CI
4	24.35	3-phenylpropyl acetate	1.45	1359	RI, MS
5	25.90	α-copaene	3.02	1378	RI, MS
6	28.40	(*E*)-caryophyllene	22.36	1420	RI, MS
7	29.50	(*E*)-cinnamyl acetate	36.59	1430	RI, MS, CI, ^{1}H NMR
8	30.28	α-humulene	5.49	1451	RI, MS
9	31.73	germacrene-D	0.53	1476	RI, MS
10	32.78	germacrene-B	0.96	1489	RI, MS
11	33.10	valencene	0.55	1490	RI, MS
12	33.23	α-muurolene	1.29	1491	RI, MS
13	33.88	γ-cadinene	2.27	1505	RI, MS
14	34.00	*cis*-γ-bisabolene	0.52	1510	RI, MS
15	34.65	δ-cadinene	4.70	1518	RI, MS
16	34.95	γ-curcumene[c]	0.56	1521	MS
17	35.28	β-guaiene[c]	0.73	1529	MS
18	36.50	ledol	2.55	1540	RI, MS
19	36.92	nerolidol	0.30	1553	RI, MS
20	37.42	spathulenol	0.40	1555	RI, MS
21	38.37	globulol	0.75	1568	RI, MS
22	39.08	10-epi-eudesmol	0.65	1581	RI, MS
23	40.03	cubenol	0.20	1610	RI, MS
24	40.83	τ-cadinol	4.90	1626	RI, MS
25	41.00	torreyol	0.65	1629	RI, MS
26	41.12	α-cadinol	1.66	1638	RI, MS
27	46.45	benzyl benzoate	1.46	1690	RI, MS, CI
			total: 95.98		

[a] KI: Kovats indices. [b] RI: retention index. MS: mass spectra. CI: co-injection with authentic sample. [c] Tentatively identified.

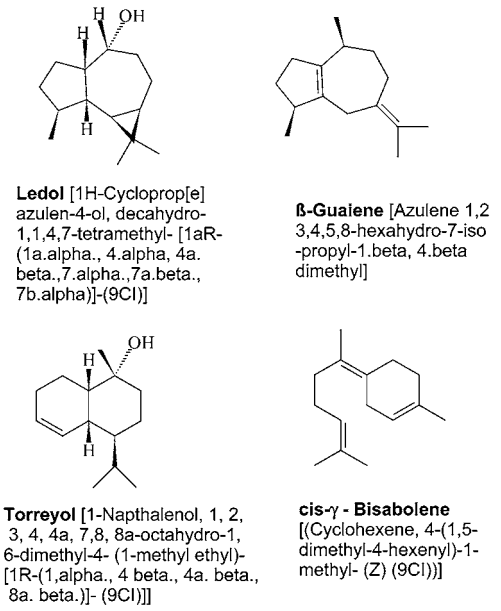

Figure 2. Structures and chemical names of four sesquiterpenes (Adams, 1989).

(39.50%), aldehydes (0.34%), and alcohols (12.76%). Several compounds represent esters and alcohols. However, aldehydes are represented by only one compound, (*E*)-cinnamaldehyde. Among the esters, (*E*)-cinnamyl acetate is the major compound and constitute to the extent of 36.59% followed by 3-phenylpropyl acetate and benzyl benzoate. All the alcohols that are present are terpenes. Acylic monoterpene alcohols were represented by only one compound, linalool. The structures and chemical names of four sesquiterpenes are presented in **Figure 2**. Among the nine sesquiterpene alcohols τ-cadinol and ledol are the major compounds and are present to the extent of 4.90% and 2.55%, respectively.

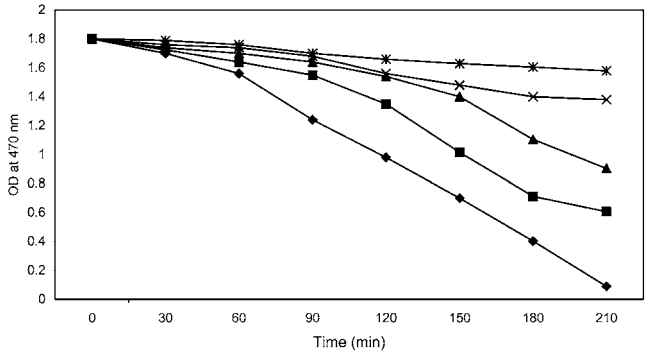

Figure 3. Antioxidant activity of cinnamon fruit stalk volatile oil and propyl gallate: ◆, control; ■, volatile oil, 100 ppm; ▲, volatile oil, 200 ppm; ×, propyl gallate, 100 ppm; and ∗, propyl gallate, 200 ppm.

Terpenic hydrocarbons are present to the extent of 42.97%. Two classes of terpenes are present, viz., monoterpenes (1.10%) and sesquiterpenes (54.51%). β-Pinene is the only monoterpene detected. Among the sesquiterpenes, β-caryophyllene is the major compound present to the extent of 20.8%, followed by δ-cadinene, α-humulene, α-copaene, γ-cadinene, α-muurolene, germacrene-B, and germacrene-D.

The antioxidant activity of cinnamon fruit stalk volatile oil and PG as measured by the bleaching of β-carotene is presented in **Figure 3**. At 200-ppm concentration, volatile oil and PG exhibit 66.9 and 92.1% antioxidant activity, respectively. The mechanism of bleaching of β-carotene is a free radical mediated phenomenon resulting from the hydroperoxides formed from linoleic acid. β-Carotene in this model system undergoes rapid discoloration in the absence of an antioxidant. The linoleic acid free radical formed upon the abstraction of a hydrogen atom from one of its diallylic methylene groups attacks the highly unsaturated β-carotene molecules. As β-carotene molecules lose their double bonds by oxidation, the compound loses its chromophore and characteristic orange color, which can be

monitored spectrophotometrically (*17*). The presence of fruit stalk volatile oil and propyl gallate can hinder the extent of β-carotene bleaching by neutralizing the linoleate free radical and other free radicals formed in the system. The activity of the volatile oil is attributed to their hydrogen-donating ability. It is well-known that free radicals cause autoxidation of unsaturated lipids in food (*19*, *20*). On the other hand, antioxidants are believed to intercept the free radical chain of oxidation and to donate hydrogen from the phenolic hydroxyl groups, thereby forming stable end product, which does not initiate or propagate further oxidation of lipid (*21*).

The phosphomolybdenum method is based on the reduction of Mo(VI) to Mo(V) by the antioxidant compounds and the formation of a green Mo(V) complex with a maximal absorption at 695 nm. The antioxidant capacity of fruit stalk volatile oil exhibited 911.2 and 1505.2 mmol/g oil (as equivalent to α-tocopherol) respectively at 100 and 200 ppm concentration.

Synthetic (*E*)-cinnamyl acetate is widely used in perfumery because of its excellent sensory and fixative properties. It is used in blossom compositions such as lilac, jasmine, and lily of the balsamic and oriental notes to the fragrances. In addition, it is used as a modifier in berry, nut, and spice flavor systems (*22*). It is also used in baked foods, meat products, soft candy, etc. (*23*). Obviously, natural (*E*)-cinnamyl acetate from fruit stalks of *C. zeylanicum* could be used for these purposes. Hence, a process has been developed for the isolation of ca. 96% pure (*E*)-cinnamyl acetate from the cinnamon fruits and flowers (*12*). The structure of the compound was further confirmed by ^{1}H NMR spectra. The signals at δ 2.1 (s, 3H) indicated the presence of a methyl group adjacent to the keto group, and δ 7.4 (s, 5H) shows the presence of five aromatic protons. The signals at δ 6.60 (d, 1H, 17 Hz) and 6.2 (dt, 1H, 17, 6 Hz) showed the presence of two trans protons and δ 4.5 (d, 6 Hz) was assigned to methylene protons.

The *C. zeylanicum* buds, flowers, fruits, and fruit stalks showed many analogies. As was reported in the previous papers, the significance of the volatile oils of four parts of cinnamon is the presence of seven compounds, which includes the major compound, i.e., (*E*)-cinnamyl acetate. Comparison of the chemical composition of volatile oils from fruit stalks, buds (*9*), flowers (*7*), and fruits (*4*) revealed that the oil from fruit stalks contains more relative amounts of shikimic acid derivatives. The character impact compound, i.e., (*E*)-cinnamyl acetate, was found to be the major compound present in the volatile oils of fruits (42−54%), flowers (42%), and fruit stalks (36.6%) whereas in the volatile oil from buds it is present to the extent of 2.4% only. Formation of this compound might have initiated in the bud stage and its further formation could have propagated during the flowering stage and reached a maximum in the fruit stage. Seventeen compounds present in the fruit volatile oil (*4*) are found in the fruit stalk oil and constitute 87.52% of the oil. The major compounds are α-copaene, δ-cadinene, (*E*)-cinnamyl acetate, α-humulene, β-caryophyllene, and τ-cadinol. Eight compounds present in the flower volatile oil (*7*) are also found in the fruit stalk oil and constitute 52.87% of the oil. These are (*E*)-cinnamaldehyde, 3-phenylpropyl acetate, (*E*)-cinnamyl acetate, α-humulene, germacrene-D, δ-cadinene, and globulol. Similarly, seven compounds represented in the bud volatile oil (*9*) are also found in the fruit stalk oil and constitute 52.54% of the oil. These are α-copaene, (*E*)-cinnamyl acetate, α-humulene, germacrene-D, δ-cadinene, globulol, and benzyl benzoate. It may be concluded that mevalonic acid metabolites formed to a maximum extent during the bud stage and increased to some extent during the fruit stage. Further, this is different from oils of other parts of *C. zeylanicum* such as leaf, root bark, and stem bark. But there are some similarities as it contains many other compounds that are present in other oils as well. It has been observed that different types of compounds are present at different stages from buds to fruits through flowers, although some of the compounds are similar.

In conclusion, *C. zeylanicum* fruit stalk volatile oil contains (*E*)-cinnamyl acetate (34.03%) and caryophyllene (22.36%) as major compounds. The former compound can be a natural substitute for the synthetic (*E*)-cinnamyl acetate used as a flavoring agent in confectioneries and liquors.

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