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Geranylated phenolic constituents from the fruits of *Artocarpus nobilis*

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Abstract

Chemical investigation of the combined dichloromethane and ethyl acetate extracts of the fruits of *Artocarpus nobilis*, furnished four new geranylated phenolic constituents, 2,4,4′-trihydroxy-3-[(2*E*)-5-methoxy-3,7-dimethylocta-2,6-dienyl]chalcone (**4**), 1-(3,4-dihydro-3,5-dihydroxy-2-methyl-2-(3-methyl-2-butenyl)-2H-1-benzopyran-6-yl-3-(4-hydroxyphenyl)-2(*E*)-propen-1-one (**5**), 8-geranyl-3′,4′,7-trihydroxyflavone (**8**), 3′-geranyl-4′,5,7-trihydroxyflavanone (**9**), together with known related compounds, xanthoangelol (**1**), xanthoangelol B (**2**), 3-geranyl-2,3′,4,4′-tetrahydroxychalcone (**3**), lespeol (**6**), 8-geranyl-4′,7-dihydroxyflavanone (**7**), and isonymphaeol-B (**10**). Compounds **3**, **8** and **10** showed strong antioxidant activity against DPPH radical by spectrophotometric method.
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Keywords: *Artocarpus nobilis*; Moraceae; Chalcones; Dihydrochalcones; Flavones; Flavonoids; Antioxidant; DPPH radical

1. Introduction

The genus *Artocarpus* of the family Moraceae is a rich source of phenolic secondary metabolites. The only endemic species of the genus *Artocarpus* growing in Sri Lanka is *Artocarpus nobilis* Thw., whose seeds and young fruits are edible. Several pyranodihydrobenzoxanthones, chromenoflavonoids, triterpenes, geranylated chalcones and stilbenes have been reported from the various parts of the plant (Pavanasasivam and Sultanbawa, 1973; Pavanasasivam et al., 1974; Kumar et al., 1977; Sultanbawa and Surendrakumar, 1989; Jayasinghe et al., 2004a,b). In a continuation of our research work on bioactive secondary metabolites from Sri Lankan plants, we carried out chemical investigation of the fruits of *A. nobilis*. In this paper we report the isolation and structure elucidation of four new geranylated phenolic metabolites (**4**, **5**, **8**, **9**) together with the known six (**1**, **2**, **3**, **6**, **7**, and **10**). Some of these compounds showed strong radical scavenging activity towards the 2,2′-diphenyl-1-picrylhydrazyl (DPPH) radical in spectrophotometric method.

2. Results and discussion

The chopped, air dried fruits of *A. nobilis* were defatted with *n*-hexane and sequentially extracted with CH_2Cl_2, EtOAc and MeOH at room temperature. None of these extracts showed significant antifungal activity against *Cladosporium cladosporioides* on TLC bioassay (Homans and Fuchs, 1970). The TLC analysis indicated that the presence of UV active compounds only in CH_2Cl_2 and EtOAc extracts. A portion of combined CH_2Cl_2 and EtOAc extracts were chromatographed over silica gel, Sephadex LH-20, reverse phase silica gel and reverse phase HPLC to give compounds **1**–**10** (Fig. 1).

Compounds **1**, **2** and **3** were identified as xanthoangelol (3-geranyl-2,4,4′-trihydroxychalcone) (**1**) (Kozawa et al., 1977), xanthoangelol B (2,4,4′-trihydroxy-3-[6-hydroxy-3,7-dimethyl-2(E),7-octadienyl]chalcone) (**2**), 3-geranyl-2,3′,4,4′-tetrahydroxy chalcone (**3**) (Baba et al., 1990) by direct comparison with the authentic samples which we previously isolated from the leaves of the same plant

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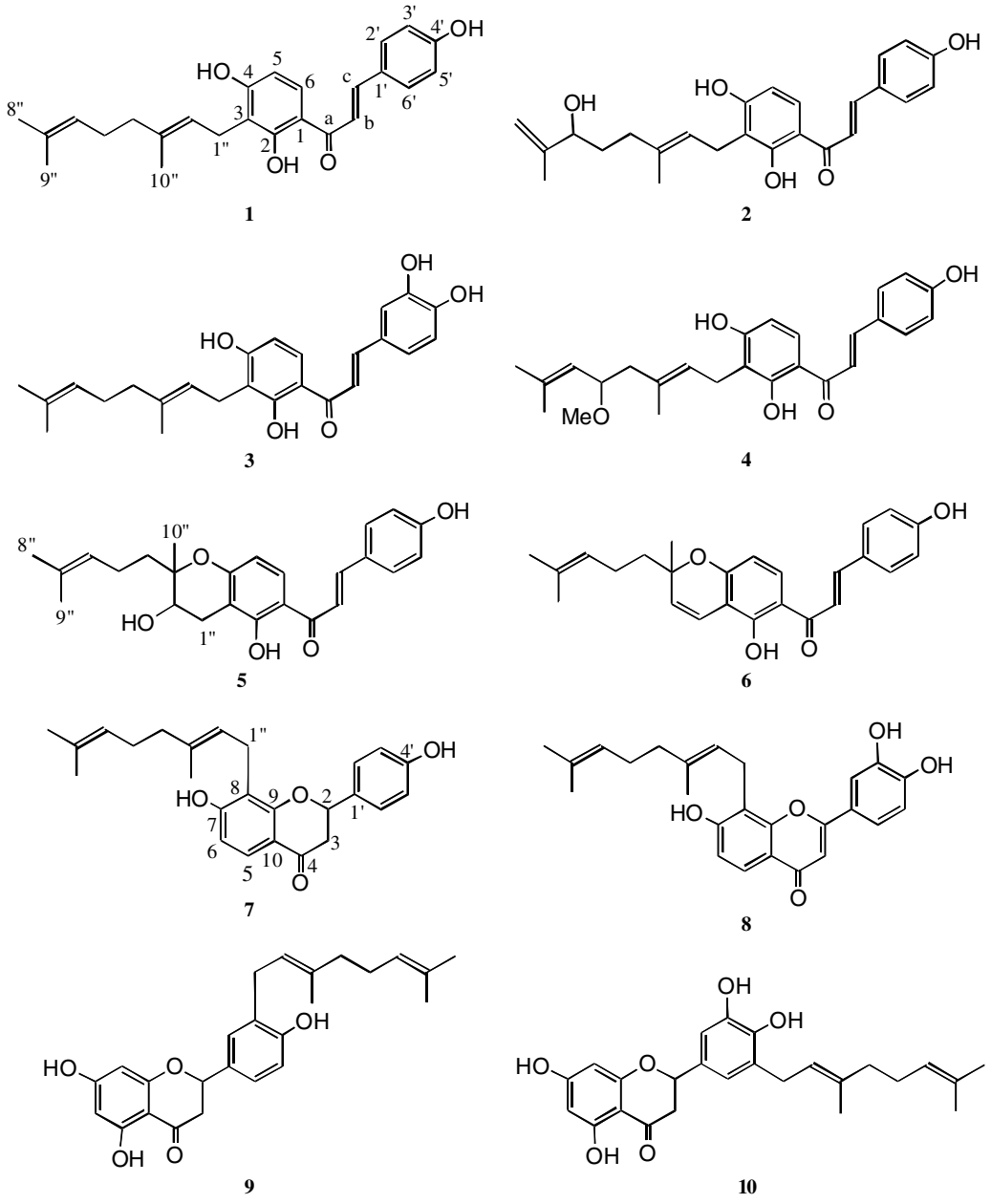

Fig. 1. Structures of compounds **1**–**10**.

(Jayasinghe et al., 2004a), while compounds **6**, **7** and **10** were determined to be lespeol (Miyase et al., 1980), prostratol F (8-geranyl-4′,7-dihydroxyflavanone) (Iinuma et al., 1995) and isonymphaeol-B (5′-geranyl-3′,4′,5,7-tetrahydroxyflavanone) (Kumazawa et al., 2004) respectively by comparison of their spectral data with those reported.

The negative FABMS spectrum of **4** indicated a peak at m/z 421[M−H]$^-$ consistent with the molecular formula $C_{26}H_{30}O_5$ {HRFABMS m/z 421.2030 (*calc.* 421.2015)}. Comparison of the ^{1}H NMR spectrum of **4** with that of **1** revealed that **4** has the same chalcone core as in **1**, showing two sets of *ortho*-coupled aromatic protons at δ 6.43 (1H, *d*, 8.9 Hz) and 7.72 (1H, *d*, 8.9 Hz) assignable to H-5 and H-6; 6.88 (2H, *d*, 8.6 Hz), 7.57 (2H, *d*, 8.6 Hz) assignable to H-3′/H-5′ and H-2′/H-6′ and two protons at δ 7.84 (*d*, 15.4) and 7.47 (*d*, 15.4) due to an α,β-unsaturated *trans*-olefin. However, the geranyl moiety at C-3 seemed to be substituted with a methoxy group (δ_H 3.25, δ_C 55.7). The HMBC spectrum revealed that the methoxy singlet was correlated with an oxymethine carbon at δ 75.2 to which a proton at δ 4.06 (*td*, $J = 8.6$, 5.7 Hz) was attached. The oxymethine proton showed correlations with an olefinic proton at δ 4.99 and methylene protons at (δ 2.15 and 2.34) in the H–H COSY spectrum. The H–H COSY spectrum further showed correlation peaks between the olefinic proton and two olefinic methyl singlets at δ 1.69 and 1.75 due to allylic coupling. These data clearly established that the methoxy group was substituted at the C-5″ position of the geranyl group. Hence, compound **4** was determined to be 2,4,4′-trihydroxy-3-[(2*E*)-5-methoxy-3,7-

dimethylocta-2,6- dienyl]chalcone. The HMBC correlations of **4** are given in Fig. 2.

The negative FABMS of **5** gave a psuedomolecular ion peak at *m/z* 407, consistent with the molecular formula $C_{25}H_{28}O_5$ {HRFABMS(−): *m/z* 407.1811 (*calc.* 407.1858). The ^{1}H NMR spectrum of **5** showed signals of the same chalcone core as in **4**, and suggested some structural variations in the geranyl moiety at C-3. The ^{13}C NMR and DEPT spectra of **5** showed new signals of a tertiary carbinyl (δ 79.4) and a secondary carbinyl (δ 67.0, associated with δ_H 3.91 (*t*, 6.4)) with disappearance of the inner double bond of the geranyl moiety. The oxymethine proton at δ 3.91 was coupled to methylene protons appeared at δ 2.97 (1H, *dd*, $J = 17.0$, 5.6 Hz) and 2.65 (1H, *dd*, $J = 17.1$, 7.2 Hz), thus assigning the oxymethine proton as H-2″ and the methylene protons as H_2-1″. ^{1}H NMR further displayed a methyl singlet signal at δ 1.34 and a set of signals due to a prenyl unit. The olefinic proton (δ 5.01 (*t*, $J = 7.0$ Hz)) of the prenyl moiety was correlated via allylic coupling to two methyl singlets at 1.60 and 1.43 and methylene protons at δ 2.13 (2H, *m*), which was further correlated with the other methylene protons at δ 1.71 (2H, *m*). Requirement of one more unsaturation suggested that compound **5** could be a higher homolog of known pyranochalcone, 1-(3,4-dihydro-3,5-dihydroxy-2,2-dimethyl-2H-1-benzopyran-6-yl-3-(4-hydroxyphenyl)-2(*E*)-propen-1-one (Abegaz et al., 2002). Comparison of the NMR data of **5** with those reported for the above gave evidence for cyclization of C-3″ and C-4 via oxygen atom to make a six-membered ring. For example, coupling constants of H_a-1″/H-2″ and H_b-1″/H-2″ (7.2 and 5.6 Hz) and chemical shift of C-1″ (δ 26.1) meet well with those of the known compound (7.0 and 5.5 Hz; δ 26.3). These data established the structure of **5** as 1-(3,4-dihydro-3,5-dihydroxy-2-methyl-2-(3-methyl-2-butenyl)-2H-1-benzopyran-6-yl-3-(4-hydroxyphenyl)-2(*E*)-propen-1-one. It can be assumed that dehydration of compound **5** would produce lespeol (**6**) in this plant.

The negative FABMS peak at *m/z* 405 $[M-H]^-$ gave evidence for the molecular formula $C_{25}H_{26}O_5$ of **8** {HRFABMS(−) *m/z*: 405.1686 (*calc.* 405.1702)}. The UV spectrum showed maxima at 258, 273, 399 nm. The ^{1}H NMR spectrum of **8** indicated an olefinic proton at δ 6.71 assignable to H-3, *ortho*-coupled two aromatic protons in ring A (δ 7.49 and 6.66), three aromatic protons due to a 3′,4′-dihydroxy B ring (δ 7.45, 6.90 and 7.39) in a flavone moiety and signals of a geranyl moiety. The chemical shifts of H-5 and H-6 as well as A-ring carbons showed good agreement with a 7-hydroxy-8-alkylatedflavone (Yin et al., 2004). An alternative structure, 6-geranyl-5-hydroxyflavone, was ruled out, since a strongly chelated OH signal expected from the structure was not observed in the ^{1}H NMR spectrum when recorded in DMSO-d_6 solvent. Hence, compound **8** was determined to be 8-geranyl-3′,4′,7-trihydroxyflavone.

Compounds **9** showed absorption maxima at 230 and 288 nm in the UV spectra. The negative FABMS spectrum of **9** gave a peak at 407 $[M-H]^-$ consistent with the molecular formula $C_{25}H_{28}O_5$ {HRFABMS(–) *m/z* 407.1824 (calc. 407.1858)}. The presence of a geranyl group was obvious from the signals of three methyl singlets at δ 1.55, 1.60 and 1.69 and two olefinic protons at δ 5.09 and 5.32 in the ^{1}H NMR spectrum. The ^{1}H NMR spectrum, further exhibited ABX type three protons [δ 2.67/3.05 (δ_c 44.0, C-3) and 5.27 (δ_c 80.6, C-2)], two *meta*-coupled aromatic protons at δ 5.87 (*d*, 2.2) assignable to H-6 and H-8 and three aromatic protons assignable to B-ring. In $CDCl_3$ solvent, a chelated OH proton was observed at δ 12.1. These data, together with a carbonyl carbon resonance at δ 197.8 (C-4), indicated a 5,7-dihydroxyflavanone structure for compound **9**. Fragment ions, *m/z* 255 and 151, arising from retro-Diels-Alder reaction in the FABMS(-) spectrum of **9** (Fig. 3) supported this formulation and further indicated that B-ring was substituted with a geranyl and a OH groups. The coupling patterns of the B-ring protons, δ 7.15 (*d*, 2.1), 6.77 (*d*, 8.2), 7.10 (*dd*, 8.2, 2.2), and HMBC correlation data (Fig. 4) established that geranyl and OH groups are located at the C-3′ and C-4′ positions, respectively. An NOE correlation from H-1″ (δ 3.29) of the geranyl moiety to H-2′ (δ 7.15) further evidenced the B-ring substitution pattern. Thus, compound **9** was established as 3′-geranyl-4′,5,7-trihydroxyflavanone.

The complete ^{1}H and ^{13}C NMR assignments of compounds **4**, **5**, **8** and **9** are given in Tables 1 and 2.

The phenolic compounds isolated in the present study, except **9** and **10**, can be biosynthetically correlated with compound **1** which has the geranyl group at the C-3 position. It would be reasonable to assume that an epoxide derivative at the internal olefin of the geranyl group of

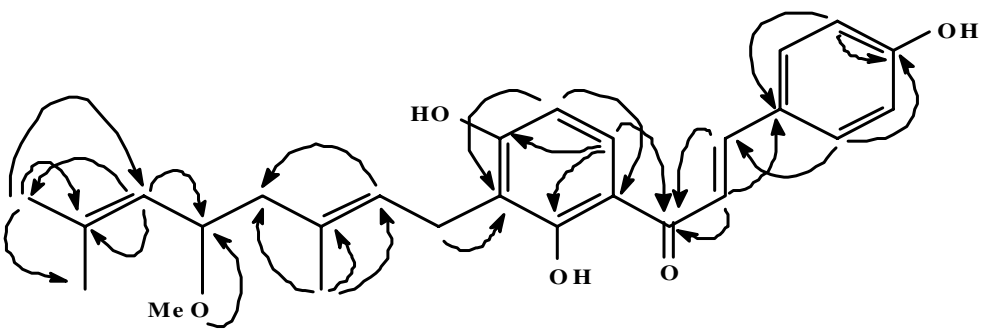

Fig. 2. HMBC correlations of **4** (H–C).

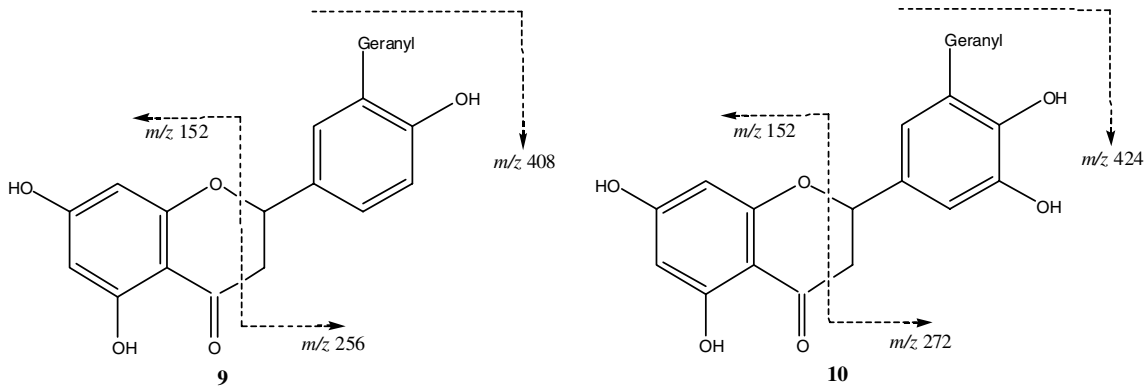

Fig. 3. FABMS (-) fragmentation pattern of **9** and **10**.

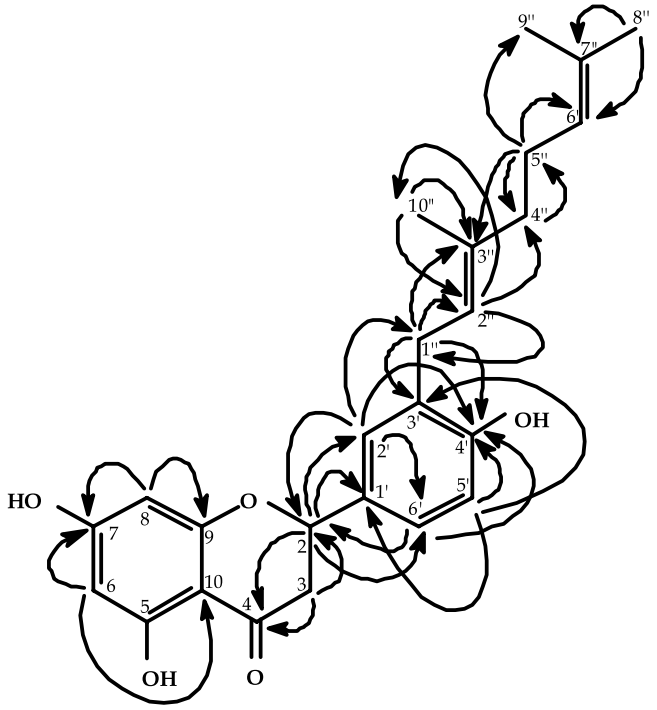

Fig. 4. HMBC correlations of **9** (H–C).

compound **3** serves as a biosynthetic precursor of **5**. An attack of the C-4 phenolic group to the epoxide could produce a pyran ring found in **5**. If this is the case, *syn*-orientation of the C-2″ hydroxyl and 10″-methyl groups is suggested, although the stereochemical study remains to be elucidated. Elimination of water from **5** could proceed without difficulty to furnish the dehydrated product **6**. One may think that compound **4** can be an artifact derived from the corresponding 5″-OH compound during extraction and isolation process. However, we are inclined to think that **4** is a real natural product, since the corresponding 5″-OH compound could not be isolated and compound **2** which has an allylic alcohol moiety failed to yield the corresponding C-6″ methoxy compound. In addition to the major metabolic pathway arising from the C-3 geranylated chalcone, the fruits have an alternative pathway which led to the C-3′ geranylated flavanones such as compounds **9** and **10**.

Antioxidant properties of all these compounds were evaluated against the DPPH radical by TLC bio-autography method (Takao et al., 1994). All these compounds **1**–**10** exhibited off-white spots in purple background at the level of 1 μg per spot. Compounds **3**, **8** and **10** showed strong off-white spots on TLC, even at the concentration of **1**–**10** reduced to 0.1 μg level. Hence, the antioxidant properties of **3**, **8** and **10** were compared with α-tocopherol using spectrophotometric method (Masuoka et al., 1997) and IC_{50} values were determined as **3** (5 μg), **8** (6.3 μg), **10** (4.4 μg), α-tocopherol (13.8 μg). Alzimer's disease, rheumatoid arthritis, cataracts, diabetes and aging itself all may be in part, caused by a phenomenon known as oxidative or free radical damage. Antioxidants are substance which can prevent, stop or reduced oxidation damage (www.aces.edu). It is well known that diet rich in fruits and vegetables significantly reduced the incidence and mortality rates of cardiovascular diseases and certain cancers in the human. Our results indicated that compounds **3**, **8** and **10** have shown high antioxidant activity in comparison with the antioxidant properties of α-tocopherol. Hence, *A. nobilis* fruits might be benefited to humans, as a rich source of natural antioxidants.

3. Experimental

3.1. General

Mps were determined by Gallenkamp apparatus and are uncorrected. Optical rotations were measured on a Perkin–Elmer 241 instrument. UV spectra were recorded on a UV-160 A spectrometer. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker DRX500 (500 MHz for ^{1}H and 125 MHz for ^{13}C) spectrometer in $CDCl_3$ or $CDCl_3$/CD_3OD (10:1) solution. Tetramethylsilane signal was used as an internal standard for ^{1}H shifts, and $CDCl_3$ ($\delta = 77.0$) signal was used as a reference for ^{13}C shifts. EIMS (70 eV) and negative FABMS were obtained on a JEOL JMS-

Table 1
^{1}H NMR (500 MHz) data of compounds **4**, **9** in $CDCl_3$ and compounds **5**, **8** in $CDCl_3/CD_3OD$ (10:1)

H No.[a]	4	5	8	9
c/2	7.84 (*d*, 15.4)	7.61 (*d*, 15.7)		5.27 (*dd*, 12.7, 3.1)
b/3	7.47 (*d*, 15.4)	7.50 (*d*, 15.7)	6.71 (*s*)	3.05 (*dd*, 17.1, 12.7)
				2.67 (*dd*, 17.1, 3.1)
2/5			7.49 (*d*, 8.4)	
3/6			6.66 (*d*, 8.4)	5.87 (*d*, 2.2)
5/8	6.43 (*d*, 8.9)	6.44 (*d*, 8.5)		5.87 (*d*, 2.2)
6/9	7.72 (*d*, 8.9)	7.52 (*d*, 8.5)		
2′	7.57 (*d*, 8.6)	7.46 (*d*, 8.7)	7.39 (*d*, 2.2)	7.15 (*d*, 2.2)
3′	6.88 (*d*, 8.6)	6.82 (*d*, 8.7)		
5′	6.88 (*d*, 8.6)	6.82 (*d*, 8.7)	6.90 (*d*, 8.3)	6.77 (*d*, 8.2)
6′	7.57 (*d*, 8.6)	7.46 (*d*, 8.7)	7.45 (*dd*, 8.3, 2.2)	7.10 (*dd*, 8.2, 2.2)
1″	3.56 (*dd*, 16.1, 6.4)	2.97 (*dd*, 17.1, 5.6)	3.54 (*d*, 7.3)	3.29 (*m*)
	3.46 (*dd*, 16.1, 8.1)	2.65 (*dd*, 17.1, 7.2)		
2″	5.38 (*dd*, 8.1, 6.4)	3.91 (*dd*, 7.2, 5.6)	5.38 (*td*, 7.3, 1.2)	5.32 (*dt*, 7.3, 4.1)
4″	2.34 (*dd*, 13.5, 8.2)	1.71 (*m*)	1.99 (*m*)	2.03 (*m*)
	2.15 (*dd*, 13.5, 5.4)			
5″	4.06 (*ddd*, 9.2, 8.2, 5.4)	2.13 (*m*)	2.01 (*m*)	2.09 (*m*)
6″	4.99 (*d*, 9.2)	5.01 (*t*, 7.0)	5.05 (*td*, 6.7, 1.4)	5.09 (*td*, 6.9, 1.3)
8″	1.75 (*s*)	1.60 (*s*)	1.60 (*s*)	1.60 (*s*)
9″	1.69 (*s*)	1.43 (*s*)	1.53 (*s*)	1.55 (*s*)
10″	1.85 (*s*)	1.34 (*s*)	1.87 (*s*)	1.69 (*s*)
MeO	3.25 (*s*)	–	–	–

[a] Chalcone/flavone numberings.

Table 2
^{13}C NMR (125 MHz) data of compounds **4**, **9** in $CDCl_3$ and compounds **5**, **8** in $CDCl_3/CD_3OD$ (10:1)

C No.	4	5	C No.	8	9
c	143.9	142.2	2	146.8	80.6
b	118.1	124.5	3	113.1	44.0
a	192.1	191.7	4	184.1	197.8
1	113.6	120.8	5	118.0	165.4
2	162.3	159.5	6	123.0	97.0
3	113.7	107.4	7	163.4	168.4
4	163.9	154.0	8	112.0	96.2
5	108.6	107.1	9	166.2	164.9
6	129.1	130.2	10	113.8	103.4
1′	127.8	127.1	1′	124.8	130.9
2′	130.5	130.1	2′	115.3	128.9
3′	116.0	115.7	3′	144.4	129.5
4′	157.9	158.9	4′	146.8	156.7
5′	116.0	115.7	5′	112.1	115.7
6′	130.5	130.1	6′	124.9	126.2
1″	22.0	26.1	1″	21.8	29.1
2″	123.7	67.0	2″	120.7	123.7
3″	136.8	79.4	3″	136.6	137.0
4″	46.0	37.5	4″	39.6	40.8
5″	75.2	21.7	5″	26.4	27.7
6″	125.4	123.6	6″	124.0	125.4
7″	136.8	132.0	7″	131.4	132.2
8″	18.3	17.3	8″	17.4	17.8
9″	25.9	25.4	9″	25.4	25.9
10″	16.4	18.3	10″	16.1	16.2
OMe	55.7	–		–	–

AX700 spectrometer. HPLC analyses were carried out on Shimadzu LC-6A apparatus equipped with UV detector using reverse phase C_{18} column under isocratic solvent condition.

3.2. Plant material

Fruits of the *A. nobilis* Thw. were collected from the central province of Sri Lanka in August, 2003. A voucher specimen is deposited at the Institute of Fundamental Studies.

3.3. Extraction and isolation

Chopped air dried whole fruit of the *A. nobilis* (800 g) were defatted with *n*-hexane and sequentially extracted with CH_2Cl_2, EtOAc and MeOH in room temperature. TLC analysis indicated that the presence of UV active spots at λ 254 nm on TLC only in the CH_2Cl_2 and EtOAc extracts in the same R_f 0.3–0.5 region (TLC eluent: 7%MeOH–$CHCl_3$). Thus only CH_2Cl_2 and EtOAc extracts were selected for further investigations. Evaporation of the combined dichloromethane extract (12 g) and ethyl acetate extracts gave light brown colored solid (14 g). A portion of combined dichloromethane and ethyl acetate extract (25 g) was chromatographed over a column of silica gel (Merck Art 7734) with *n*–hexane–EtOAc–MeOH followed by a combination of chromatographies over column of silica gel, Sephadex LH-20, reverse phase silica gel and reverse phase HPLC [STR Prep ODS 20 × 250 mm column, 5 ml/min, 20% H_2O–MeOH, UV detection 254 nm] furnished compounds **1** (12 mg), **2** (8 mg), **3** (10 mg), **4** (4 mg), **5** (11 mg), **6** (6 mg), **7** (11 mg), **8** (7 mg), **9** (80 mg), **10** (24 mg).

2,4,4′-trihydroxy-3-[(2E)-5-methoxy-3,7-dimethylocta-2,6-dienyl]chalcone (**4**). Yellow colored amorphous; $[\alpha]_D^{25} = 0^\circ$ (*c* 0.1 in MeOH); UV_{max}^{MeOH} (log ε): 225 (sh) (4.1), 310 (sh) (3.8), 368 (4.3); FABMS(–): *m*/*z* 421 $[M–H]^-$

{HRFABMS(–): *m/z* 421.2030 (*calc*. 421.2015 for $C_{26}H_{29}O_5$)}.

1-(3,4-dihydro-3,5-dihydroxy-2-methyl-2-(3-methyl-2-butenyl)-2H-1-benzopyran-6-yl-3-(4-hydroxyphenyl)-2(E)-propen-1-one (**5**). Yellow colored amorphous; $[\alpha]_D^{25} = -48^\circ$ (*c* 0.31 in MeOH); UV_{max}^{MeOH} (log ε): 230(4.2), 356(4.4); FABMS(–): *m/z* 407[M−H]$^-$. {HRFABMS(–): *m/z* 407.1811 (*calc*. 407.1858 for $C_{25}H_{27}O_5$}.

8-geranyl-3′,4′,7-trihydroxyflavone (**8**). M.p. 113 °C; UV_{max}^{MeOH} (log ε): 258 (3.9), 273 (3.9), 399 (4.2); FABMS-(–): *m/z* 405 [M–H]$^-$ {HRFABMS(–): *m/z* 405.1686 (*calc*. 405.1702 for $C_{25}H_{25}O_5$)}.

3′-geranyl-4′,5,7-trihydroxyflavanone (**9**). Yellow colored amorphous; UV_{max}^{MeOH} (log ε): 230 (sh) (4.2), 288 (4.0); FABMS(–): *m/z* 407[M–H]$^-$ {HRFABMS (–): *m/z* 407.1824 (*calc.* 407.1858 for $C_{25}H_{27}O_5$)}.

3.4. Antioxidant activity

The antioxidant activity was evaluated by the DPPH radical scavenging effect. The 0.1 M acetate buffer was prepared by dissolving 1.64 g of CH_3COONa in 16 ml of H_2O and 150 μl of CH_3COOH. The final volume was adjusted to 20 ml by adding H_2O. The 0.2 mM DPPH solution was prepared by dissolving 3.9 mg of DPPH in 50 ml of ethanol. α-Tocopherol (1 mg) in 10 ml of ethanol solution was prepared. A series of test tubes with 1.0 ml of buffer solution was mixed well with 0.5 ml of DPPH solution. A series of various concentrations of compounds **3**, **8**, **10** and α-tocopherol (1 μg–20 μg in 1 ml of ethanol) were added to each tube and mixed well. After 30 min in room temperature the absorptions of each solution was measured by UV spectrophotometer at 517 nm. A mixture of buffer solution and ethanol was used as the reference for the spectrophotometer. A graph was plotted with the weight of the compound vs. absorptions and IC_{50} values determined. The antioxidant activity was expressed in terms of IC_{50} (μg/ml, concentration required to inhibit the DPPH radical formation by 50%). α-Tocopherol was used as a positive control.

References

Abegaz, B.M., Ngadjui, B.T., Dongo, E., Ngamemi, B., Nindi, M.N., Bezabih, M., 2002. Chalcones and other constituents of *Dorstenia prorepens* and *Dorstenia zenkeri*. Phytochemistry 59, 877–883.

Baba, K., Nakata, K., Taniguchi, M., Kido, T., Kozawa, M., 1990. Chemical components of *Angelica keiskei*. Part 7. Chalcones from *Angelica keiskei*. Phytochemistry 29, 3907–3910.

Homans, A.L., Fuchs, A., 1970. Direct bioautography on thin-layer chromatograms as a method for detecting fungitoxic substances. Journal of Chromatography 51, 327–329.

Iinuma, M., Ohyama, M., Tanaka, T., 1995. Flavonoids in roots of *Sophora prostrata*. Phytochemistry 38, 539–543.

Jayasinghe, L., Balasooriya, B.A.I.S., Padmini, W.C., Hara, N., Fujimoto, Y., 2004a. Geranyl chalcone derivatives with antifungal and radical scavenging properties from the leaves of *Artocarpus nobilis*. Phytochemistry 65, 1287–1290.

Jayasinghe, U.L.B., Puvanendran, S., Hara, N., Fujimoto, Y., 2004b. Stilbene derivatives with antifungal and radical scavenging properties from the stem bark of *Artocarpus nobilis*. Natural Product Research 18, 571–574.

Kozawa, M., Morita, N., Baba, K., Hata, K., 1977. The structure of xanthoangelol, a new chalcone from the roots of *Angelica keiskei* Koidzumi (Umbelliferae). Chemical & Pharmaceutical Bulletin 25, 515–516.

Kumar, N.S., Pavanasasivam, G., Sultanbawa, M.U.S., Mageswaran, R., 1977. Chemical investigation of Ceylonese plants. Part 24. New chromenoflavonoids from the bark of *Artocarpus nobilis* Thw. (Moraceae). J.C.S. Perkin Transactions 1 (11), 1243–1251.

Kumazawa, S., Goto, H., Hamasaka, T., Fukumoto, S., Fujimoto, T., Nakayama, T., 2004. A new prenylated flavonoid from propolis collected in Okinawa, Japan. Bioscience, Biotechnology, and Biochemistry 68, 260–262.

Masuoka, C., Ono, M., Ito, Y., Nohara, T., 1997. Antioxidative constituents from the aerial part of *Piper elongatum* Vahl. Food Science and Technology International, Tokyo 3, 285–289.

Miyase, T., Ueno, A., Noro, T., Fukushima, S., 1980. Studies on the constituents of *Lespedeza cyrtobotrya* Miq. I. The structures of a new chalcone and two new isoflav-3-ens. Chemical & Pharmaceutical Bulletin 28, 1172–1177.

Pavanasasivam, G., Sultanbawa, M.U.S., 1973. Chemical investigation of Ceylonese plants. VI. Cycloartenyl acetate, cycloartenol and cycloartenone in the bark of *Artocarpus* species. Phytochemistry 12, 2725–2726.

Pavanasasivam, G., Sultanbawa, M.U.S., Mageswaran, R., 1974. Chemical investigation of Ceylonese plants. X. New chromenoflavonoids from the bark of *Artocarpus nobilis* (Moraceae). Chemistry and Industry 21, 875–876.

Sultanbawa, M.U.S., Surendrakumar, S., 1989. Two pyranodihydrobenzoxanthones from *Artocarpus nobilis*. Phytochemistry 28, 599–605.

Takao, T., Kitatani, F., Watanabe, N., Yagi, A., Sakata, K., 1994. A simple screening method for antioxidant produced by marine bacteria from fish and shellfish. Bioscience, Biotechnology and Biochemistry 58, 1780–1783.

Yin, S., Fan, C.-K., Wang, Y., Dong, L., Yue, J.-M., 2004. Antibacterial prenylflavone derivatives from *Psoralea corylifolia*, and their structure–activity relationship study. Bioorganic & Medicinal Chemistry 12, 4387–4392.