ISSN 1859 - 4735

JOURNAL OF MEDICINAL MATERIALS

No. 1+2 - Vol. 31
3/2026

SPECIAL ISSUE

THE 14th SCIENTIFIC CONFERENCE OF NATIONAL INSTITUTE OF MEDICINAL MATERIALS (NIMM)
THE 65th ANNIVERSARY OF NIMM FOUNDATION (13/4/1961-13/4/2026)

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CHEMICAL CONSTITUENTS FROM THE PROPOLIS OF THE STINGLESS BEE *LEPIDOTRIGONA TERMINATA (SMITH, 1878)* COLLECTED IN GIA LAI

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Received January 26th, 2026 Accepted March 7th, 2026

Summary

Six compounds, including gracilol A (**1**), dipterocarpol (**2**), 8-hydroxylabdan-15-oic acid (**3**), (*E*)-1-(2-methoxy-4′,5′-dihydroxyphenyl)-3-phenylprop-2-ene (**4**), (+)-eudesmin (**5**), and (-)-*epi*-eudesmin (**6**), were isolated from propolis of stingless bee *Lepidotrigona terminata* (Smith, 1878) collected in Van Duc, Gia Lai province. The structures of isolated compounds **1**−**6** were elucidated by spectroscopic (NMR, MS) analysis and comparison with previous literature. This is the first report on the NMR data of compounds **1** and **4**.

Keywords: *Lepidotrigona terminata; Triterpene; Diterpene; Lignan; Phenolic.*

1. Introduction

Propolis is a complex resinous substance collected by bees from plant exudates and mixed with salivary secretions and wax. Propolis has been used since ancient times for its diverse therapeutic properties. While propolis of the honeybee (*Apis mellifera*) has been extensively studied, garnering significant scientific and commercial interest, propolis produced by stingless bees remains underexplored [1],[2]. Stingless bee propolis, known variably as "cerume" or geopropolis, differs in botanical origins, physical properties, and chemical profiles from that of honeybee (*A. mellifera*). The chemical composition of stingless bee propolis is remarkably diverse and geographically variable, reflecting the specific flora foraged by different meliponine species [2]. Phytochemical investigations have revealed a diverse profile of compounds, including phenolic acids, flavonoids, terpenoids, and lignans, many of which are unique to specific bee-plant interactions [3],[4],[5]. Pharmacological studies of propolis extracts and their isolated compounds exhibit a wide range of biological activities such as antibacterial, anticancer, antioxidant, and anti-inflammatory effects [2].

Lepidotrigona terminata (Smith, 1878) is a stingless bee species widely distributed across Southeast Asia, including Thailand, Malaysia, Vietnam, and Indonesia [6]. Nevertheless, relatively few studies have characterized the chemical composition and bioactivity of *L. terminata* propolis. The *L. terminata* propolis extract in Thailand, containing xanthone compounds, displayed weak cytotoxic, antioxidant, and anti-diabetic activities [7],[8]. Using the GC/MS method, Popova et al. reported that *L. terminata* propolis samples in Vietnam had cardanols, fatty acids, and triterpenes with antibacterial effect [9]. There has been no isolation study of natural compounds from *L. terminata* propolis in Vietnam so far. In our ongoing research on natural compounds and bioactive substances of stingless bee propolis, we report herein our first investigation on chemical constituents from *L. terminata* propolis collected in Van Duc, Gia Lai province.

2. Materials and methods

2.1. Materials

The propolis sample was collected from a stingless bees hive in beekeeping house at Van Duc town, Gia Lai province, Vietnam, in 2024. The stingless bee species was identified as *Lepidotrigona terminata* (Smith, 1878) by Prof. Nguyen Thi Phuong Lien and Dr. Tran Thi Ngat, Institute of Biology, Vietnam Academy of Science and Technology. Voucher specimens were deposited at the Department of Insect Ecology, Institute of Biology, Vietnam Academy of Science and Technology.

2.2. General experimental procedures

ESI-MS spectra were recorded on an Agilent 1260 series single quadrupole LC/MS system (Agilent, CA, USA). NMR spectra (^{1}H, ^{13}C, HSQC, HMBC, and NOESY) were obtained on

Bruker AVANCE III HD 500 MHz or Bruker AVANCE NEO 600 MHz spectrometers (Bruker, Billerica, MA, USA) with tetramethylsilane as the internal standard. Column chromatography (CC) was performed on normal phase *silica gel* 60 (230-400 mesh, Merck) and Sephadex LH20. Thin layer chromatography was performed on *silica gel* 60 F_{254} and RP-18 $F_{254}S$. Compounds were visualized by spraying with 10% sulfuric acid in EtOH and heating.

2.3. Extraction and isolation

The propolis sample (500 g) was macerated and extracted with 90% EtOH (4 times × 5 L for 48 h) at room temperature. The combined extracts were concentrated under reduced pressure to yield 254 g of a dark brown EtOH extract. The EtOH extract (250 g) was suspended in 800 mL of distilled water and then partitioned with EtOAc (4 × 500 mL). The combined EtOAc extracts were evaporated under reduced pressure to afford the EtOAc extract (165.7 g). The EtOAc residue (160 g) was applied to *silica gel* CC, eluted with a solvent gradient of *n*-hexane–EtOAc (100:1 to 0:100) to yield 12 fractions (E1-E12). Fraction E3 (63.0 g) was chromatographed by normal phase *silica gel* CC (*n*-hexane–EtOAc, 19:1, v/v) to give 11 fractions (E3.1-E3.11). Fraction E3.6 (1.8 g) was separated by *silica gel* CC (*n*-hexane–acetone, 6:1, v/v) to yield ten fractions E3.6.1-E3.6.10. Fraction E3.6.1 (85.3 mg) was then purified by *silica gel* CC (*n*-hexane–EtOAc, 8:2, v/v) to afford **3** (7.0 mg). Fraction E3.9 (450 mg) was subjected to *silica gel* CC (*n*-hexane–acetone, 19:1, v/v), yielding 11 sub-fractions E3.9.1-E3.9.11. Fraction E3.9.6 (50 mg) was further purified by *silica gel* CC (*n*-hexane–acetone, 19:1, v/v) to give **1** (3.6 mg). Fraction 3.10 (100 mg) was separated by *silica gel* CC (*n*-hexane–acetone, 19:1, v/v) to afford **2** (15.6 mg). Fraction E8 (8.96 g) was separated on a normal-phase *silica gel* column, eluted with a gradient solvent system of *n*-hexane–EtOAc (7:1 to 3:1), yielding 12 sub-fractions (E8.1 to E8.12). Fraction E8.7 (2.98 g) was chromatographed on normal-phase *silica gel* CC (*n*-hexane–acetone *v/v*, 9:1), to afford **4** (3.6 mg). Fraction E8.10 (2.98 g) was purified on normal-phase *silica gel* CC (CH_2Cl_2–EtOAc, 9:1, v/v), and then subsequently on Sephadex LH20 CC (MeOH) to give **6** (5.4 mg). Fraction E8.11 (197 mg) was purified on Sephadex LH20 CC (MeOH) to yield **5** (10.3 mg).

Gracilol A (1): white solid; ESI-MS: *m/z* 433 $[M+H]^+$; ^{1}H-NMR (600 MHz, $CDCl_3$) and ^{13}C-NMR (125 MHz, $CDCl_3$): see Table 1.

Dipterocarpol (2): white solid; ESI-MS *m/z* 433 $[M+H]^+$; ^{1}H-NMR (500 MHz, $CDCl_3$) and ^{13}C-NMR (125 MHz, $CDCl_3$): see Table 1.

8-Hydroxylabdan-15-oic acid (**3**): yellow solid; ESI-MS: *m/z* 323 $[M-H]^-$; ^{1}H-NMR (600 MHz, $CDCl_3$) and ^{13}C-NMR (125 MHz, $CDCl_3$): see Table 2.

(E)-1-(2'-Methoxy-4',5'-dihydroxyphenyl)-3-phenylprop-2-ene (4): red oil; ^{1}H-NMR (600 MHz, $CDCl_3$) and ^{13}C-NMR (125 MHz, $CDCl_3$): see Table 2.

(+)-Eudesmin (5): yellow solid; $[\alpha]_D^{25}$ + 48° (*c* 0.5, MeOH); ESI-MS *m/z* 387 $[M+H]^+$; ^{1}H-NMR (500 MHz, CD_3OD): δ_H 7.00 (2H, s, H-2, H-2'), 6.94 (4H, m, H-5, H-5', H-6, H-6'), 4.76 (2H, d, *J* = 4.5 Hz, H-7, H-7'), 4.26 (2H, m, H-9a, H-9'a), 3.88 (2H, dd, *J* = 3.5, 9.0 Hz, H-9b, H-9'b), 3.85 (6H, s, 2 × OMe), 3.83 (6H, s, 2 × OMe), 3.16 (2H, m, H-8, H-8'); ^{13}C-NMR (125 MHz, CD_3OD): δ_C 150.7 (C-3, C-3'), 150.2 (C-4, C-4'), 135.3 (C-1, C-1'), 119.8 (C-6, C-6'), 113.0 (C-5, C-5'), 111.3 (C-2, C-2'), 87.3 (C-7, C-7'), 72.7 (C-9, C-9'), 56.6 (2 × OMe), 56.5 (2 × OMe), 55.4 (C-8, C-8').

(-)-epi-Eudesmin (6): pale yellow solid; $[\alpha]_D^{25}$ -85° (*c* 0.5, $CHCl_3$); ESI-MS *m/z* 387 $[M+H]^+$; ^{1}H-NMR (600 MHz, $CDCl_3$): δ_H 6.94 (1H, br s, H-2), 6.92 (1H, d, *J* = 1.8 Hz, H-2'), 6.90 (1H, dd, *J* = 1.8, 8.4 Hz, H-6), 6.86 (1H, dd, *J* = 1.8, 8.4 Hz, H-6′), 6.85 (1H, d, *J* = 8.4 Hz, H-5′), 6.84 (1H, d, *J* = 8.4 Hz, H-5), 4.88 (1H, d, *J* = 5.4 Hz, H-7′), 4.45 (1H, d, *J* = 7.2 Hz, H-7), 4.14 (1H, d, *J* = 10.2 Hz, H-9a), 3.91 (3H, s, OMe), 3.90 (3H, s, OMe), 3.89 (3H, s, OMe), 3.88 (3H, s, OMe), 3.86 (2H, m, H-9′a, H-9b), 3.33 (2H, m, H- H-9′b, H-8′), 2.92 (1H, m, H-8); ^{13}C-NMR (125 MHz, CD_3OD): δ_C 149.3 (C-3), 148.9 (C-3′), 148.8 (C-4), 148.1 (C-4′), 133.7 (C-1), 131.0 (C-1′), 118.5 (C-6), 117.8 (C-6′), 111.1 (C-5,C-5′), 109.3 (C-2), 109.1 (C-2′), 87.7 (C-7), 82.1 (C-7′), 71.1 (C-9), 69.8 (C-9′), 56.0 (2 × OMe), 55.98 (OMe), 55.95 (OMe), 54.5 (C-8), 50.2 (C-8′).

3. Results and discussion

The ethyl acetate extract of the propolis of the stingless bee *L. terminata* was separated using combined chromatographic methods to afford six compounds, including gracilol A (**1**), dipterocarpol (**2**), 8-hydroxylabdan-15-oic acid (**3**), (*E*)-1-(2′-methoxy-4′,5′-dihydroxyphenyl) -3-phenylprop-2-ene (**4**), (+)-eudesmin (**5**), and (-)-*epi*-eudesmin (**6**) (Fig. 1).

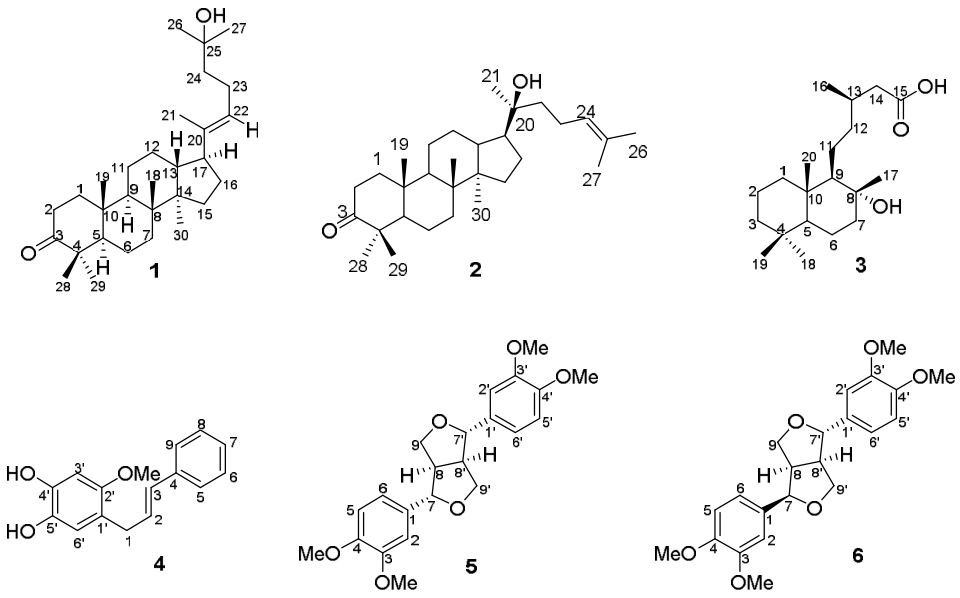

Fig. 1. Chemical structures of compounds **1**–**6** from *L. terminata* propolis

Compound **1** was isolated as a white solid. The ^{1}H-NMR spectrum of **1** exhibited characteristic signals of a triterpene with 8 methyl singlets at δ_H 1.00 (3H, s, H-18), 0.94 (3H, s, H-19), 1.56 (3H, s, H-21), 1.23 (6H, s, H-26, H-27), 1.08 (3H, s, H-28), 1.04 (3H, s, H-29), and 0.86 (3H, s, H-30). In addition, an olefinic proton was observed at δ_H 5.15 (1H, t, J = 7.2 Hz, H-22). The ^{13}C-NMR and HSQC spectra of **1** showed 30 carbons including a carbonyl carbon at the downfield δ_C 218.9 (C-3); 2 olefinic carbons at δ_C 137.3 (C-20) and 124.5 (C-22), an oxygenated carbon at δ_C 71.2 (C-25); eight methyl groups at δ_C 15.3 (C-18), 16.1 (C-19), 13.0 (C-21), 29.2 and 29.3 (C-26 and C-27), 26.8 (C-28), 21.0 (C-29), and 15.8 (C-30). The ESI-MS spectrum of **1** showed a protonated molecular ion peak at *m/z* 433 $[M+H]^+$, which gave evidence of its molecular formula as $C_{30}H_{50}O_2$ (M = 432), corresponding to six DBE. The NMR and MS spectra suggested that **1** was a tetracyclic triterpene. The ketone group at C-3 was confirmed by the HMBC correlations of H-28 (δ_H 1.08) and H-29 (δ_H 1.04) to C-3 (δ_C 218.9). The tertiary alcohol substituted group at C-25 was determined by the HMBC correlations of H-26 and H-27 (δ_H 1.23) to C-25 (δ_C 71.2). The HMBC cross-peaks of H-22 (δ_H 5.15) to C-21 (δ_C 13.0) and C-17 (δ_C 50.3), of H-21 (δ_H 1.56) to C-20 (δ_C 137.3) and C-22 (δ_C 124.5) suggested the location of the double bond at C-20 and C-22 (Fig. 2). The *E* configuration of the double bond was assigned based on NOESY cross-peaks of H-22 (δ_H 5.15) with H-17 (δ_H 2.17), and of H-21 (δ_H 1.56) with H-23 (δ_H 2.08). The NOESY spectrum also confirmed the dammarane structure. NOESY cross-peaks of H-30 (δ_H 0.86) with H-17 (δ_H 2.17) and H-9 (δ_H 1.41) suggested that the H-30 methyl group and protons H-9 and H-17 were on the same side. Compound **1** was identified as gracinol A. This compound was reported from *Dipterocarpus gracilis*, but the data could not be accessed [10]. The NMR data of this compound were documented for the first time.

Compound **2** was isolated as a white solid. The molecular formula of **2** was also assigned as $C_{30}H_{50}O_2$ based on a protonated molecular ion peak at *m/z* 433 $[M+H]^+$ in the ESI-MS spectrum and NMR data. The ^{1}H- and ^{13}C-NMR spectra of **2** revealed similar signals to those of **1**. In the ^{1}H-NMR spectrum, eight methyl singlets [δ_H 1.00 (3H, s, H-18), 0.93 (3H, s, H-19), 1.15 (3H, s, H-21), 1.69 (3H, s, H-26), 1.62 (3H, s, H-27), 1.08 (3H, s, H-28), 1.04 (3H, s, H-29), and 0.89 (3H, s, H-30)], and an olefinic proton [δ_H 5.12 (1H, t, J = 6.0 Hz, H-24)] were detected. The ^{13}C NMR and DEPT also exhibited 30 carbon signals including a ketone group [δ_C 218.0 (C-3)], 2 olefinic carbon signals [δ_C 124.7 (C-24) and 131.6 (C-25)], an oxygenated carbon [δ_C 75.3 (C-20)] and 8 methyl groups [δ_C 15.2 (C-18), 16.0 (C-19), 25.5 (C-21), 25.7 (C-26), 17.7 (C-27), 26.7 (C-28), 21.0 (C-29), and 16.3 (C-30)]. The NMR data suggested that **1** and **2** are structural isomers. Compound **2** was assigned as dipterocarpol by comparison with previously reported data [11].

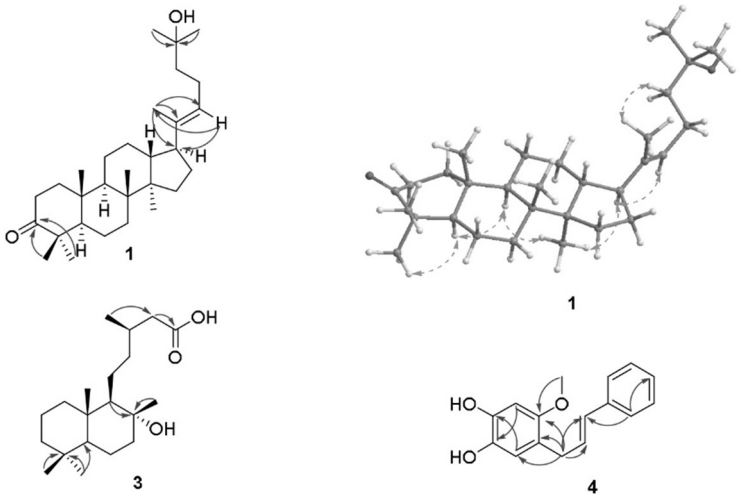

Fig. 2. Key HMBC (⟶) and NOESY (⟷) correlations of compounds **1**, **3**, and **4**

Table 1. ^{1}H- and ^{13}C-NMR spectroscopic data of compounds **1** and **2**

Pos.	**1**		**2**		**Ref [11]**	
	δ_H[a] (mult., *J* in Hz)	δ_C[b]	δ_H[c] (mult., *J* in Hz)	δ_C[b]	δ_H[c] (mult., *J* in Hz)	δ_C[b]
1	1.93 (m)/1.46 (m)	40.0		39.9		39.9
2	2.49-2.43 (m)	34.1	2.42-2.49 (m)	34.1	2.37-2.42 (m)	34.1
3		218.9		218.0		218.0
4		47.4		47.4		47.4
5	1.39 (m)	55.4		55.3		55.4
6	1.55-1.46 (m)	19.7		19.6		19.7
7	1.61 (m)/1.33 (m)	34.8		34.5		34.6
8		40.4		40.3		40.3
9	1.41 (m)	50.4*		50.0		50.0
10		37.0		36.8		36.8
11	1.52 (m)/1.26 (m)	22.0		22.0		22.0
12	1.80 (m)/1.42 (m)	27.3		27.5		27.5
13	1.62 (m)	44.5		42.4		42.4
14		49.2		50.2		50.3
15	1.52 (m)/1.11 (m)	31.6		31.1		31.2
16	1.60 (m)/1.47 (m)	24.9		24.8		24.8
17	2.17 (m)	50.3*		49.8		49.8
18	1.00 (s)	15.3	1.00 (s)	15.2	0.93 (s)	15.2
19	0.94 (s)	16.1	0.93 (s)	16.0	0.88 (s)	16.0
20		137.3		75.3		75.4
21	1.56 (s)	13.0	1.15 (s)	25.5	1.08 (s)	25.5
22	5.15 (t, 7.2 Hz)	124.5		40.5		40.5
23	2.08 (m)	23.0	2.05 (m)	22.6	1.99 (m)	22.6
24	1.53 (m)	43.7	5.12 (t, 6.0)	124.7	5.05 (t, 5.0)	124.7
25		71.2		131.6		131.6
26	1.23 (s)	29.2**	1.69 (s)	25.7	1.62 (s)	25.7
27	1.23 (s)	29.3**	1.62 (s)	17.7	1.56 (s)	17.7
28	1.08 (s)	26.8	1.08 (s)	26.7	1.01 (s)	26.7
29	1.04 (s)	21.0	1.04 (s)	21.0	0.97 (s)	21.0
30	0.86 (s)	15.8	0.89 (s)	16.3	0.82 (s)	16.4

*[a]: $CDCl_3$, 600 MHz, [b]: $CDCl_3$, 125 Mz [c]: $CDCl_3$, 500 MHz, *, **: the signals can be exchanged.*

Compound **3** was isolated as a white solid. The ESI-MS spectrum showed the peak *m/z* 323 $[M–H]^-$, suggesting the molecular formula $C_{20}H_{36}O_3$ (DBE = 3). The ^{1}H-NMR spectrum of **3** displayed signals for 5 methyl groups including 4 singlets [δ_H 1.15 (3H, s, H-17), 0.83 (3H, s, H-18), 0.86 (3H, s, H-19), and 0.75 (3H, s, H-20)] and 1 doublet [δ_H 0.98 (3H, d, *J* = 6.6 Hz, H-16)]. The ^{13}C-NMR and HSQC spectra showed signals for 20 carbons, including 5 methyl groups at δ_C 19.7 (C-16), 30.6 (C-17), 33.4 (C-18), 21.7 (C-19), and 15.1 (C-20); 8 methylene groups at δ_C 38.9 (C-1), 18.2 (C-2), 41.8 (C-3), 18.3 (C-6), 40.7 (C-

7), 22.6 (C-11), 42.2 (C-12), and 42.1 (C-14); 3 methine groups at δ_C 56.0 (C-5), 59.3 (C-9), and 31.2 (C-13); 2 quaternary carbons not bearing hydrogen at δ_C 33.2 (C-4), and 39.2 (C-10); 1 oxygenated carbon at δ_C 73.2 (C-8); and 1 carboxylic acid group at δ_C 173.2 (C-15). The HMBC spectrum allowed the assignment of proton and carbon signals. HMBC correlations of H-18 (δ_H 0.83) and H-19 (δ_H 0.86) to C-4 (δ_C 33.2) and C-5 (δ_C 56.0); of H-17 (δ_H 1.15) and H-9 (δ_H 1.12) correlated to C-8 (δ_C 73.2); and the HMBC signal of H-14 (δ_H 2.12 and 2.31) correlated to C-15 (δ_C 173.2) suggested the labdane skeleton. Comparing the NMR data with reference [12], compound **3** was determined as 8-hydroxylabdan-15-oic acid.

Table 2. ^{13}C-NMR spectroscopic data of compounds **3** and **4**

C	3		Ref [12]		C	4	
	δ_C[a]	δ_H[b] (mult. *J* in Hz)	δ_C[c]	δ_H[d] (mult. *J* in Hz)		δ_C[a]	δ_H[b]
1	38.9		39.8		1	32.6	3.50 (d, 6.6)
2	18.2		18.6		2	129.0	6.32 (dd, 15.6, 6.6)
3	41.8		41.5		3	130.9	6.43 (d, 15.6)
4	33.2		33.4		4	137.5	-
5	56.0		56.3		5	126.1	7.33 (d, 7.8)
6	18.3		20.6		6	128.5	7.28 (t, 7.8)
7	40.7		40.7		7	127.1	7.19 (t, 7.8)
8	73.2		75.1		8	128.5	7.28 (t, 7.8)
9	59.3	1.12 (s)	62.3		9	126.1	7.33 (d, 7.8)
10	39.2		39.3		1'	124.6	-
11	22. 6		23.2		2'	148.5	-
12	42.2		44.1		3'	111.3	6.67 (s)
13	31.2		31.4		4'	136.1	-
14	42.1	2.12 (m)/2.31 (m)	42.2		5'	143.1	-
15	173.2		178.0		6'	121.1	6.67 (s)
16	19.7	0.98 (d, 6.6 Hz)	20.2	0.98 (d, 6.6 Hz)	OMe	61.4	3.82 (s)
17	30.6	1.15 (s)	24.1	1.14 (s)			
18	33.4	0.83 (s)	33.4	0.83 (s)			
19	21.7	0.86 (s)	21.7	0.85 (s)			
20	15.1	0.75 (s)	15.7	0.77 (s)			

[a:] $CDCl_3$, 600 MHz, [b:] $CDCl_3$, 125 MHz, [c:] $CDCl_3$, 300 MHz, [d:] $CDCl_3$, 750 MHz.

Compound **4** was isolated as a red oil. The ^{1}H-NMR spectrum of **4** showed signals for 9 protons in the aromatic/double bond region, including 5 protons of a phenyl group at δ_H 7.33 (2H, d, *J* = 7.8 Hz, H-5 and H-9), 7.28 (2H, t, *J* = 7.8 Hz, H-6 and H-8), and 7.19 (1H, t, *J* = 7.8 Hz, H-7); two aromatic proton singlets at δ_H 6.67 (2H, s, H-3' and H-6'); and two *trans*-protons of a double bond at δ_H 6.43 (1H, d, *J* = 15.6 Hz, H-3) and 6.32 (1H, dd, *J* = 15.6, 6.6 Hz, H-2). Additionally, there is a methoxy group signal at δ_H 3.82 (3H, s, OMe) and a methylene group signal at δ_H 3.50 (2H, d, *J* = 6.6 Hz, H-1). The ^{13}C-NMR and HSQC spectra showed signals for 16 carbons, with 14 signals in the region for double-bond or aromatic carbons. The first phenyl ring included signals at δ_C 137.5 (C-4), 126.1 (C-5, C-9), 128.5 (C-6, C-8), and 127.1 (C-7). The second phenyl ring showed carbon signals at δ_C 124.6 (C-1'), 148.5 (C-2'), 111.3 (C-3'), 136.1 (C-4'), 143.1 (C-5'), and 121.1 (C-6'). The signals for carbons in the propene side chain are at δ_C 130.9 (C-3), 129.0 (C-2), and 32.6 (C-1). In addition, the methoxy group was at δ_C 61.4 (OMe). In the HMBC spectrum, correlations of H-5 and H-9 (δ_H 7.33) to C-3 (δ_C 130.9), of H-2 (δ_H 6.32) to C-4 (δ_C 137.5) indicate the linkage C-3/C-4. HMBC correlations of H-1 (δ_H 3.50) to C-1' (δ_C 124.6), C-6' (δ_C 121.1), and C-2' (δ_C 148.5) suggested the 1,2,4,5-substitutions at the second phenyl ring. The HMBC correlations of H-3' and H-6' (δ_H 6.67) to C-4' (δ_C 136.1) and C-5' (δ_C 143.1) confirmed the substitution pattern. The correlation of methoxy protons (δ_H 3.82) to C-2' (δ_C 148.5) confirmed the methoxy group at C-2'. Based on the spectral analysis, **4** is determined to be (*E*)-1-(2'-methoxy-4',5'-dihydroxyphenyl)-3-phenylprop-2-ene. Compound **4** was found in *Dalbergia retusa* (Fabaceae family), but the authors only reported the NMR data of a diacetylated product [13],[14]. Therefore, this is the first time that we report the NMR spectral data of this compound, as well as its first discovery in stingless bee propolis. Compound **4** has a structure similar to compound (*Z*)-1-(2'-methoxy-4',5'-dihydroxyphenyl)-3-phenylprop- 2-ene, found in red Mexican propolis [15].

Compounds **5** and **6** were determined as (+)-

eudesmin (**5**) and (-)-*epi*-eudesmin (**6**), respectively, based on the comparison of NMR data with those in the previous reports [16],[17].

This is the first chemical study of stingless bee *L. terminata* (Smith, 1878) propolis in Vietnam. Compound **2** was identified from propolis of *Lisotrigona furva* [3] and *Lepidotrigona terminata* [9]. Compound **5** was isolated from the propolis of *Lepidotrigona ventralis* collected in Tuyen Quang province [18]. To the best of our knowledge, compounds **1**, **3**, **4**, and **6** were first reported from stingless bee propolis. Notably, gracinol A (**1**) and dipterocarpol (**2**) have been found in the plants *Dipterocarpus* spp. [10], while compound **4** was only isolated from *Dalbergia retusa* [13],[14]. Based on our chemical results, resin sources for *L. terminata* propolis could be *Dipterocarpus* spp. and *Dalbergia retusa* plants. Our findings are in agreement with those of Popova et al., that *Lepidotrigona* sp. propolis displayed complicated and diverse chemical composition due to multiple resin sources [9].

4. Conclusions

Phytochemical study of stingless bee *Lepidotrigona terminata* propolis resulted in six constituents, including gracilol A (**1**), dipterocarpol (**2**), 8-hydroxylabdan-15-oic acid (**3**), (*E*)-1-(2′-methoxy-4′,5′-dihydroxyphenyl)-3-phenylprop-2-ene (**4**), (+)-eudesmin (**5**), and *epi*-eudesmin (**6**). Compounds **1**, **3**, **4**, and **6** were identified for the first time from stingless bee propolis. NMR spectral data of compounds **1** and **4** were first documented. The plants *Dipterocarpus* sp. and *Dalbergia retusa* are proposed as the resin sources for *L. terminata* propolis in Gia Lai province.

Acknowledgements: *The study was funded by the Gia Lai Department of Science and Technology.*

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