# Chemotaxonomic Relationship of Oligomer Resveratrol in Three Malaysian *Dipterocarpus* Species from the Taxonomic Tribe of Dipterocarpaceae

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**Abstract:** A phytochemical investigation of three species of Malaysian Dipterocarpus contributed to the isolation of 22 compounds which consist of 15 oligostilbenoids, 2 terpenes, 2 coumarins, and 3 flavonoids. The isolation of flavonoids in the Dipterocarpaceae family is very limited. Moreover, 4-methoxepigallocatechin-3-O-O-(3-methyl) gallate (20) was isolated for the first time in the plant. The occurrence of 4-O-methylgallocatechin (18) and its stereoisomer; 4-O'-methylepigallocatechin (19) was first reported in the Dipterocarpaceae family. This study also reported the existence of several types of oligostilbenoids such as davidiol A (8), stenophyllol B (9), isohopeaphenol (11), resveratrol (1), and ampelopsin E (10) which are the first occurrence in Dipterocarpus, more closely to Vatica which is classified under Dipterocarpeae tribe.

*Keywords: chemotaxonomy; Dipterocarpaceae;* Dipterocarpus; *flavonoid; oligostilbenoids* 

# INTRODUCTION

*Dipterocarpus* is one of the main genera of Dipterocarpaceae, which consists of 75 species. This genus is the third largest genera in the Dipterocarpaceae family, after *Shorea* (150 species) and *Hopea* (100 species) [1]. Despite its significance in the family, *Dipterocarpus* has been the subject of limited research. The chemical properties of various *Dipterocarpus* species have been investigated, revealing the presence of resveratrol oligomers and triterpenoids [2]. Besides, ursolic acid, quercetin, and catechin were isolated from *Dipterocarpus retusus* [3]. There are only 9 species that were repeatedly isolated as resveratrol oligomers, which are *D. hasseltii* [4], *D. retusus* [4], *D. grandiflorus* [5], *D. verrucosus* [6], *D. cornutus* [7], *D. intricatus* [8], *D. semivestitus* [9-10], and *D. alatus* [11].

*D. alatus* was also used to harvest triterpenoid [12]. Table 1 shows the constituents of chemical properties of seven genera of tribe Dipterocarpeae, which are *Dipterocarpus* (9 species), *Vatica* (12 species), *Upuna* (1 species), *Anisoptera* (3 species), *Stemonoporous* (1 species), *Vateria* (2 species), and *Cotylelobium* (2 species) [13]. *Dipterocarpus, Cotylelobium, Anisoptera,* and *Stemonoporous* genera showed the ability to produce resveratrol oligomers up to tetramer. In contrast, *Vatica* sp. produced a higher degree of polymerization, which is up to hexamer, heptamer and octamer. Meanwhile, *Upuna* produced up to hexamer, and *Vateria* produced up to octamer resveratrol.

Additionally, the phylogenetic placement of *Dipterocarpus* species within the Dipterocarpoideae sub-

family remains unclear, indicating the need for further research in this area [14]. Numerous studies have been done to clarify the controversy regarding the number of genera of the Dipterocarpoideae subfamily. In addition, Sri Lankan Dipterocarpus species (D. glandulosus, D. hispidus, D. insignis, and D. zeylanicus) form a separate clade and Dryobalanops also form a distinct and highly supported monophyletic clade [15]. A study done by Cvetković et al. [16] provides strong support for revising classification the tribal of the subfamily of Dipterocarpoideae into four main clades: Dipterocarpeae (Dipterocarpus), Dryobalanopseae (Dryobalanops), Shoreeae (Hopea, Neobalanocarpus, Parashorea, and all parts of a polyphyletic Shorea) and Vaterieae (including all other presently accepted Dipterocarpoideae genera). A study reveals Hopea forms a clade with Shorea sections Anthoshorea and Doona [17]. Meanwhile, Dipterocarpus is placed as a sister to the tribe Shoreae. This separates Dipterocarpus from the remaining genera of tribe Dipterocarpeae containing the following genera: Anisoptera, Cotylelobium, Stemonoporus, Upuna, Vateria, Vateriopsis and Vatica [18]. The inconsistency of placement of Dipterocarpus in molecular phylogenies is consistent with its unique morphology, Cvetković et al. [16] and Ashton and Heckenhauer [18] proposed to isolate it in a monotypic tribe, requiring the renaming of the former Tribe Dipterocarpeae: Dipterocarpeae, Vaticeae and Shoreae.

The classification of Asian Dipterocarps into taxonomic relevant units (tribes, genera, sections, subsections) has been reviewed by Widians et al. [19] based on the previous work by Aslam et al. [1] and others [20-22]. Furthermore, the chemotaxonomy of *Dipterocarpus* and its relationship with other genera in Dipterocarpaceae have been explored, shedding light on the chemical constituents of different genera within the tribe Dipterocarpeae.

Studies have also identified the potential biological activities of *Dipterocarpus* species, such as antidiabetic and antiplasmodial properties [23]. Moreover, the effects of *Dipterocarpus* species, such as *Dipterocarpus alatus*, on UV B-protection, collagen stimulation, and nitric oxide inhibition have been investigated [24]. The bioactivity of

secondary metabolites from Dipterocarpus species are antidiabetic, antiplasmodial, antibacterial, antioxidant, anti-classes, cytotoxic, anticholinesterase, antiproliferation, anti-inflammatory and antimicrobial [25]. Fractions isolated from *D. intricatus* flowers can be utilized as natural antimicrobial, antioxidant, and cytotoxic agents for medicine [26]. The Keruing wood contained extractive substances with the main compound of bioactive caryophyllene and the total caryophyllene content in extractive wood reached 47.68% [27].

Recent researchers have conducted numerous studies on resveratrol due to its highly promising bioactivities [28-32] and its most prominent stilbenoid synthesized by plants [33]. Resveratrol demonstrated a significant effect in formulations for dermatology and cosmetics [34-35], a promising candidate for the development of nutraceuticals and pharmaceuticals [36], modulates the inflammatory response [37] and drug formulation [38]. Resveratrol dimers such as ε-viniferin exhibited strong activities against inflammatory and oxidative stress [39]. Higher degree of resveratrols such α-viniferin possess potential antidiabetic and antiplasmodial activities [40], meanwhile, (-)hopeaphenol showed its potential in inhibiting the viral entry across multiple SARS-CoV-2 variants [41]. These findings underscore the importance of further research to fully understand the chemical composition and biological activities of Dipterocarpus species.

# EXPERIMENTAL SECTION

#### Materials

Samples of the stem bark of *D. verrucosus*, *D. crinitus*, and *D. cornutus* were collected in March 2010 from the forest reserve UiTM Jengka, Pahang, Malaysia. The plants were identified by a botanist, and a voucher specimen (SKD1, SKD2, and SKD3) was deposited in the herbarium of Universiti Teknologi MARA, Malaysia (Pahang Campus).

#### Instrumentation

Infrared (IR) spectra were recorded on the Spectrum One FTIR spectrometer (Perkin-Elmer). The ultraviolet (UV) spectra were recorded on a UV-vis 160i (Shimadzu). The optical rotation was measured on the Autopolar VI Automatic Polarimeter. The melting points (uncorrected) were determined using a micro-melting point apparatus. HRESI-MS spectra were obtained with Agilent Technologies 6224 TOF LC/MS. The 1D- and 2D-NMR data were obtained from FT Bruker 300 Ultra shield (300 MHz for <sup>1</sup>H and 75 MHz for <sup>13</sup>C), JEOL UKM (500 MHz for <sup>1</sup>H and 100 MHz for <sup>13</sup>C), JEOL Meijo Nagoya University Pharm Japan (500 MHz for <sup>1</sup>H and 100 MHz for <sup>13</sup>C), and Bruker 500 Ultra shield (500 MHz for <sup>1</sup>H and 100 MHz for <sup>13</sup>C) (RIND UiTM) using various commercially available deuterated solvents such as chloroform- $d_{4}$ , acetone- $d_{6}$ , and methanol- $d_{4}$ . Mestrenova software was used to analyze the spectrum in detail. The vacuum liquid chromatography (VLC) was carried out using Si-gel Merck 60 GF254 (230-400 mesh, cat No. 1.07747), the process of column chromatography (CC) was performed with Si-gel Merck 60 (200-400 mesh), Sephadex LH<sub>20</sub>, and thin layer chromatography (TLC) analysis on pre-coated Si-gel plate with Si-gel Merck Kieselgel 60 F254 0.25 mm, 20 × 20 cm, cat No 1.05554, and radial chromatography with Merck Si-gel 60 GF 254 (5-40 µm, cat. No 1.07749).

#### Procedure

The stem barks of *D. verrucosus* were cut into small pieces, air-dried, and ground into fine powder. The finely ground plant materials were weighed (6 kg) and macerated with acetone ( $4 \times 9$  L). The acetone extract was concentrated to a volume of 250 mL. Diethyl ether was added to the concentrated acetone extract to obtain ethersoluble and insoluble fractions that are free from tannin. The soluble material was evaporated in a vacuum at 40 °C to yield 60 g crude extract. The extract was stored at room temperature. The isolation process started with  $2 \times 30$  g crude extract using VLC with a 10 cm in diameter column and silica gel weighed 250 g. This crude was chromatographed by n-hexane (Hex)-ethyl acetate (EtOAc), ethyl acetate-methanol (MeOH) to methanol (100%) (gradience of increasing methanol) to provide five fractions (DV1-DV5). The fractions were subjected to further isolation using repeated VLC and were purified by repeated RC, CC, and PTLC on silica gel, eluted with various solvent systems such as chloroform (CHCl<sub>3</sub>)-

MeOH, Hex-CHCl<sub>3</sub>-MeOH, CHCl<sub>3</sub>-Hex, and CHCl<sub>3</sub>-EtOAc-MeOH. The same procedure above was repeated on the samples of *D. cornutus* (5 kg) and *D. crinitus* (4 kg).

From the study, isolation using repeated VLC and purification by repeated RC, CC, and PTLC on the stem barks of *D. verrucosus* discovered 9 compounds. The compound consists of 8 oligostilbenes and 1 phenolic compound. Fraction 2 attained laevifonol (**3**) (10 mg) and  $\varepsilon$ -viniferin (**2**) (6 mg), Fraction 3 found ampelopsin E (**10**) (9 mg),  $\alpha$ -viniferin (**6**) (15 mg), and vaticanol B (**13**) (7 mg). In addition, fraction 4 found diptoindonesin E (**14**) (8 mg), while fraction 5 attained isohopeaphenol (**12**), (hopeaphenol) (**13**) (20 mg), and 1 non-oligostilbenoid namely bergenin (**16**) (15 mg).

Meanwhile, the extraction of *D. cornutus* successfully isolated 10 compounds consisting of 6 oligostilbenoids, 3 catechins, and 1 coumarin. Fraction 2 found scopoletin (17) (17 mg), davidiol A (8) (15 mg), stenophyllol B (9) (15 mg), and laevifonol (3) (40 mg). Additionally, fraction 3 attained  $\varepsilon$ -viniferin (2) (8 mg), fraction 4 attained ampelopsin F (4) (7 mg), and fraction 5 yielded 4-O-methylgallocatechin (18) (15 mg), 4-O-metylgallocatechin (19) (12 mg) and new compounds, which were 4-methoxy epigallocathechin-3-O-(3-methyl) gallate (20) (15 mg) and hemsleyanol D (15) (15 mg).

Additionally, the *D. crinitus* extraction efficiently isolated 8 compounds, including 5 oligostilbenoids, 2 terpenoids, and 1 phenolic compound. The *D. crinitus* extraction efficiently isolated 8 compounds, including 5 oligostilbenoids, 2 terpenoids, and 1 phenolic compound. Fraction 2 attains  $\beta$ -sitosterol (**21**) (10 mg) and  $\beta$ -sitosterol glucoside (**22**) (13 mg). Meanwhile, fraction 3 found resveratrol (**1**) (10 mg) and  $\varepsilon$ -viniferin (**2**) (9 mg). In addition, fraction 4 successfully isolated vaticanol A (7) (7 mg), ampelopsin A (**5**) (10 mg),  $\alpha$ viniferin (**6**) (7 mg), and bergenin (**16**) (8 mg). Fig. 1 shows all the isolated compounds.

# RESULTS AND DISCUSSION

In this study, 15 resveratrol oligomers from *D. verrucosus*, *D. cornutus*, and *D. crinitus* which consist of 1 monomer (resveratrol), 4 dimers; [ $\epsilon$ -viniferin (2), laevifonol (3), ampelopsin A (5), ampelopsin F (4)], 5 trimers; [ $\alpha$ -viniferin (6), vaticanol A (7), davidiol A (8), stenophyllol B (9), ampelopsin E (10)] and 5 tetramers; [isohopeapenol (11), hopeapenol (12), vaticanol B (13), diptoindonesin E (14), hemsleyanol D (15)] have been isolated and identified (Fig. 1).

**Resveratrol (1)**, obtained as an amorphous white crystal. m.p.: 220–224 °C (dec.).  $[\alpha]_D$ : +100° (*c* 0.1, MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 229, 315 nm. <sup>1</sup>H-NMR (acetone-*d*<sub>6</sub>, 500 MHz)  $\delta_H$  ppm: 7.40 (2H, *d*, *J* = 8.4, H-2a/6a), 6.83 (2H, *d*, *J* = 9.0, H-3a/5a), 7.02 (1H, *d*, *J* = 16.5, H-7a), 6.86 (1H, *d*, *J* = 16.5, H-8a), 6.54 (2H, *d*, *J* = 2.2, H-10a/14a), 6.27 (1H, *d*, *J* = 2.0, H-12a).<sup>13</sup>C-NMR (125 MHz)  $\delta_C$  ppm: 128.7 (C-1a), 129.7 (C-2a/6a), 116.4 (C-3a/5a), 158.2 (C-4a), 129.1 (C-7a), 126.8 (C-8a), 140.8 (C-9a), 105.7 (C-10a), 159.4 (C-11a), 102.7(C-12a), 159.6 (C-13a).

ε-Viniferin (2), obtained as a brownish viscous oil, MS m/z: 455 [MH]<sup>+</sup>. m.p.: 172–176 °C.  $[\alpha]_D^{20}$ : -44° (c 0.1 MeOH). UV (MeOH) λ<sub>max</sub>: 203, 230, 324 nm. IR (KBr) v<sub>max</sub> (cm<sup>-1</sup>): 3383 (OH), 1640, 1514, 1440 (C=C aromatic), and 832 (para-disubstituent). <sup>1</sup>H-NMR (methanol-d<sub>4</sub>, 300 MHz)  $\delta_{\rm H}$  ppm: 7.18 (2H, *d*, *J* = 8.7, H-2a/6a), 6.81 (2H, *d*, *J* = 8.7, H-3a/5a), 5.39 (1H, *d*, *J* = 6.6, H-7a), 4.35 (1H, *d*, *J* = 6.6, H-8a), 6.18 (2H, d, J = 1.8, H-10a/14a), 6.20 (1H, d, d)*J* = 2.1, H-12a), 7.07 (2H, *d*, *J* = 8.7, H-2b/6b), 6.68 (2H, *d*, *J* = 8.7, H-3b/5b), 6.87 (1H, *d*, *J* = 16.2, H-7b), 6.61 (1H, *d*, *J* = 16.2, H-8b), 6.27 (1H, *d*, *J* = 1.8, H-12b), 6.65 (1H, *d*, *J* = 1.8, H-14b). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 132.8 (C-1a), 127.8 (C-2a/6a), 115.3 (C-3a/5a), 158.7 (C-4a), 93.0 (C-7a), 56.1 (C-8a), 146.6 (C-9a), 106.1 (C-10a), 160.0 (C-11a), 101.2 (C-12a), 160.0 (C-13a), 106.1 (C-14a), 129.1 (C-1b), 127.0 (C-2b/6b), 115.4 (C-3b/5b), 157.3 (C-4b), 122.3 (C-7b), 129.2 (C-8b), 135.5 (C-9b), 118.9 (C-10b), 161.6 (C-11b), 96.1 (C-12b), 161.6 (C-13b), 103.3 (C-14b).

**Laevifonol (3)**, obtained as a white crystal, m.p.: 298– 300 °C.  $[\alpha]_D^{20}$ : -175° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 226, 284 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3364 (OH), 2913 (C–H), 1614, 1587, 1516, 1454, 1440 (C=C aromatic) and 835 (*para*-disubstituent), 1257 (aryl-O), 1789 (C=O). <sup>1</sup>H-NMR (methanol-*d*<sub>4</sub>, 300 MHz)  $\delta_H$  ppm: 6.98 (2H, *d*, *J* = 8.7, H-2a/6a), 6.7 7(2H, *d*, *J* = 8.7, H-3a/5a), 5.29 (1H, *d*, *J* = 10.8, H-7a), 3.29 (1H, *d*, *J* = 10.8, H-8a), 6.20 (1H, *d*, *J* = 2.0, H-12a), 7.14 (1H, *brs*, H-14a), 6.77 (2H, *d*, *J* = 8.1, H-2b/6b), 6.77 (2H, *d*, *J* = 8.1, H-3b/5b), 5.06 (1H, *d*, *J* = 7.5, H-7b), 3.29 (1H, *d*, *J* = 10.5, H-8b), 5.92 (2H, *d*, *J* = 2.1, H-10b/14b), 6.16 (1H, *t*, *J* =2.1, H-12b), 4.41 (1H, *brs*, H-4'), 4.21 (1H, *m*, H-5'), 3.97 (1H, *dd*, *J* = 4.4, H-6'). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 128.3 (C-1a), 127.4 (C-2a/6a), 115.2 (C-3a/5a), 157.5 (C-4a), 89.0 (C-7a), 55.2 (C-8a), 127.5 (C-9a), 122.0 (C-10a), 160.4 (C-11a), 95.9 (C-12a), 158.2 (C-13a), 109.8 (C-14a), 131.2 (C-1b), 129.0 (C-2b/6b), 115.0 (C-3b/5b), 157.8 (C-4b), 93.4 (C-7b), 55.3 (C-8b), 130.2 (C-9b), 131.1 (C-10b), 121.7 (C-11b), 95.9 (C-12b), 157.9 (C-13b), 110.1 (C-14b).

Ampelopsin F (4), obtained as white crystal, m.p.: 218-220 °C. MS *m*/*z*: 455 [MH<sup>+</sup>]. [α]<sub>D</sub><sup>20</sup>: +60° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}\!\!:$  203, 226, 284 nm. IR (KBr)  $\nu_{max}$ (cm<sup>-1</sup>): 3364 (OH), 2913 (C-H), 1614, 1587, 1516, 1454, 1440 (C=C aromatic) and 835 (para-disubstituent). <sup>1</sup>H-NMR (acetone- $d_6$ , 600 MHz)  $\delta_H$  ppm: 7.09 (2H, d, J =8.4, H-2a/6a), 6.76 (2H, d, J = 8.4, H-3a/5a), 4.18 (1H, d, *J* = 1.5, H-7a), 3.35 (1H, *brs*, H-8a), 6.06 (1H, *d*, *J* = 2.4, H-12a), 6.54 (1H, *d*, *J* = 2.4, H-14a), 6.78 (2H, *d*, *J* = 8.4, H-2b/6b), 6.58 (2H, d, J = 8.4, H-3b/5b), 3.64 (1H, brs, H-7b), 4.12 (1H, *brs*, H-8b), 6.15 (1H, *d*, *J* = 2.1, H-12b), 6.48 (1H, d, J = 2.4, H-14b). <sup>13</sup>C-NMR (150 MHz)  $\delta_{\rm C}$ ppm: 138.5 (C-1a), 129.8 (C-2a/6a), 115.6 (C-3a/5a), 156.3 (C-4a), 47.2 (C-7a), 58.4 (C-8a), 147.3 (C-9a), 127.8 (C-10a), 153.3 (C-11a), 101.9 (C-12a), 158.7 (C-13a), 104.1 (C-14a), 135.4 (C-1b), 129.7 (C-2b/6b), 115.7 (C-3b/5b), 156.4 (C-4b), 50.5 (C-7b), 49.7 (C-8b), 147.2 (C-9b), 113.4 (C-10b), 157.9 (C-11b), 101.9 (C-12b), 157.3 (C-13b), 105.7 (C-14b).

**Ampelopsin A (5)**, obtained as a yellow crystal. MS *m/z*: 469 [MH<sup>-</sup>]. m.p.: 218–220 °C. [α]<sub>D</sub><sup>20</sup>: –160° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 226, 284 nm. IR (KBr)  $\nu_{max}$ (cm<sup>-1</sup>): 3364 (OH), 2913 (C–H), 1614, 1587, 1516, 1454, 1440 (C=C aromatic) and 835 (*para*-disubstituent). <sup>1</sup>H-NMR (acetone-*d*, 500 MHz)  $\delta_{\rm H}$  ppm: 7.11 (2H, *d*, *J* = 8.6, H-2a/6a), 6.75 (2H, *d*, *J* = 8.7, H-3a/5a), 5.75 (1H, *d*, *J* = 11.5, H-7a), 4.15 (1H, *brs*, H-8a), 6.42 (1H, *d*, *J* = 2.3, H-10a,), 6.22(1H, *d*, *J* = 2.3, H-12a), 6.89 (2H, *d*, *J* = 8.0, H-2b/6b), 6.63 (2H, *d*, *J* = 8.8, H-3b/5b), 5.44 (1H, *d*, *J* = 4.6, H-7b), 5.40 (1H, *d*, *J* = 4.6, H-8b), 6.14 (1H, *d*, *J* = 2.0, H-12b), 6.64 (1H, *d*, *J* = 2.0, H-14b). <sup>13</sup>C-NMR (125 MHz)



1





















Fig 1. Structure of compounds isolated from D. verrucosus, D. cornutus, and D. crinitus

 $\delta_{\rm C}$  ppm: 132.7 (C-1a), 129.9 (C-2a/6a), 116.0 (C-3a/5a), 158.5 (C-4a), 88.5 (C-7a), 49.6 (C-8a), 143.6 (C-9a), 118.4 (C-10a), 157.3 (C-11a), 101.6 (C-12a), 158.9 (C-13a), 105.6 (C-14a), 131.0 (C-1b), 128.8 (C-2b/6b), 115.4 (C-3b/5b), 156.1 (C-4b), 43.9 (C-7b), 71.2 (C-8b), 140.5 (C-9b), 118.9 (C-10b), 160.2 (C-11b), 97.1 (C-12b), 158.9 (C-13b), 110.5 (C-14b).

**a-Viniferin (6)**, obtained as pale yellow, MS *m/z*: 677 [MH<sup>-</sup>]. m.p.: 220–223 °C.  $[\alpha]_D^{20}$ : +60° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 226, 284 nm. IR (KBr)  $v_{max}$  (cm<sup>-1</sup>): 3393 (OH), 1613, 1462, 1337 (C=C aromatic), and 831 (*para*-disubstituent). <sup>1</sup>H-NMR (acetone-*d*<sub>6</sub>, 300 MHz)  $\delta_H$  ppm: 7.02 (2H, *d*, *J* = 8.7, H-2a/6a), 6.71 (2H, *d*, *J* = 8.7, H-3a/5a), 6.08 (1H, *s*, H-7a), 3.97(1H, *brs*, H-8a), 6.00 (1H, *d*, *J* = 2.1, H-12a), 6.23 (1H, *d*, *J* = 2.1, H-14a), 7.22 (2H, *d*,

*J* = 8.7, H-2b/6b), 6.79 (2H, *d*, *J* = 8.7, H-3b/5b), 5.96 (1H, *d*, *J* = 9.9, H-7b), 4.71 (1H, *d*, *J* = 9.9, H-8b). 6.73 (1H, *d*, *J* = 2.1, H-12b), 6.25 (1H, *d*, *J* = 2.1, H-12b), 7.06 (2H, *d*, *J* = 8.7, H-2c/6c), 6.80 (2H, *d*, *J* = 8.7, H-3c/5c), 4.91 (1H, *d*, *J* = 6.3, H-7c), 4.61 (1H, *d*, *J* = 6.3, H-8c), 6.60 (1H, *d*, *J* = 1.8, H-12c), 6.22 (1H, *d*, *J* = 2.1, H-14a), <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 132.0 (C-1a), 128.1 (C-2a/6a), 115.7 (C-3a/5a), 157.8 (C-4a), 86.4 (C-7a), 46.4 (C-8a), 118.8 (C-9a), 141.2 (C-10a), 159.3 (C-11a), 108.5 (C-12a), 161.5 (C-13a), 98.0 (C-14a), 132.2 (C-1b), 128.6 (C-2b/6b), 116.1 (C-3b/5b), 158.2 (C-4b), 89.9 (C-7b), 52.8 (C-8b), 120.9 (C-9b), 139.7 (C-10b), 159.34 (C-11b), 106.2 (C-12b), 158.4 (C-13b), 96.8 (C-14b), 132.4 (C-1c), 128.6 (C-2c/6c), 116.0 (C-3c/5c), 158.2 (C-4c), 95.5 (C-7c), 55.6 (C-8c), 119.6 (C-9c), 138.6 (C-10c),

160.9 (C-11c), 105.7 (C-12c), 161.7 (C-13c), 96.8 (C-14c). Vaticanol A (7), obtained as a brown amorphous powder. MS m/z: 681 [MH<sup>-</sup>]. m.p.: 230–233 °C.  $[\alpha]_D^{20}$ : -90° (c 0.1 MeOH). UV (MeOH) λ<sub>max</sub>: 203, 226, 284 nm. IR (KBr) v<sub>max</sub> (cm<sup>-1</sup>): 3418 (OH), 1614, 1515, 1455 (C=C aromatic) and 833 (para-disubstituent). <sup>1</sup>H-NMR (methanol- $d_4$ , 300 MHz)  $\delta_{\rm H}$  ppm: 7.22 (2H, d, J = 8.7, H-2a/6a), 6.82 (2H, *d*, *J* = 8.7, H-3a/5a), 6.15 (1H, *d*, *J* = 3.0, H-7a), 4.37 (1H, d, J = 3.0, H-8a), 5.97 (1H, d, J = 2.4, H-12a), 6.48 (1H, *d*, *J* = 2.4, H-14a), 7.06 (2H, *d*, *J* = 8.7, H-2b/6b), 6.61 (2H, *d*, *J* = 8.7, H-3b/5b), 5.10 (1H, *d*, *J* = 10.0, H-7b), 4.53 (1H, *d*, *J* = 6.6, H-8b), 6.27 (1H, *brs*, H-12b), 6.50(2H, *d*, *J* = 8.7, H-2c/6c), 6.37 (2H, *d*, *J* = 8.7, H-3c/5c), 3.62 (1H, *d*, *J* = 7.2, H-7c), 4.23 (1H, *s*, H-8c), 6.37 (1H, *d*, *J* = 2.1, H-10c), 6.24 (1H, *t*, *J* = 2.1, H-11c), 6.37 (1H, *d*, *J* = 2.1, H-12c). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 133.7 (C-1a), 126.8 (C-2a/6a), 115.1 (C-3a/5a), 154.9 (C-4a), 85.6 (C-7a), 49.2 (C-8a), 144.0 (C-9a), 118.4 (C-10a), 156.9 (C-11a), 100.3 (C-12a), 156.6 (C-13a), 101.1 (C-14a), 137.6 (C-1b), 128.3 (C-2b/6b), 114.2 (C-3b/5b), 157.8 (C-4b), 35.0 (C-7b), 46.8 (C-8b), 144.2 (C-9b), 118.9 (C-10b), 157.8 (C-11b), 94.3 (C-12b), 157.8 (C-13b), 122.3 (C-14b), 135.3 (C-1c), 128.8 (C-2c/6c), 113.9 (C-3c/5c), 154.8 (C-4c), 63.8 (C-7c), 56.2 (C-8c), 146.6 (C-9c), 106.0 (C-10c), 158.7 (C-11c), 99.8 (C-12c), 158.7 (C-13c), 106.0 (C-

Davidiol A (8), obtained as a brown amorphous powder. MS *m/z*: 679 [MH<sup>-</sup>]. m.p.: 255–257 °C. [a]<sub>D</sub><sup>20</sup>: –275° (c 0.1 MeOH). UV (MeOH) λ<sub>max</sub>: 203, 226, 284 nm. IR (KBr) v<sub>max</sub> (cm<sup>-1</sup>): 3418 (OH), 1614, 1515, 1455 (C=C aromatic), and 833 (para-disubstituent). <sup>1</sup>H-NMR (acetone- $d_{6}$ , 300 MHz)  $\delta_{\rm H}$  ppm: 7.21 (2H, *d*, *J* = 8.7, H-2a/6a), 6.80 (2H, *d*, *J* = 8.7, H-3a/5a), 6.09 (1H, *d*, *J* = 3.0, H-7a), 4.42 (1H, *d*, *J* = 9.6, H-8a), 6.44 (1H, *d*, *J* = 2.1, H-12a), 6.57 (1H, *d*, *J* = 2.4, H-14a), 7.02 (2H, *d*, *J* = 8.7, H-2b/6b), 6.60 (2H, *d*, J = 8.7, H-3b/5b, 5.28 (1H, br s, H-7b), 4.27 (1H, d, J =11.4, H-8b). 6.04 (1H, s, H-12b), 6.74 (2H, d, J = 8.7, H-2c/6c), 6.61 (2H, *d*, *J* = 8.7, H-3c/5c), 4.39 (1H, *d*, *J* = 9.3 H-7c), 2.93 (1H, *dd*, *J* = 11.7, 9.9, H-8c), 6.43 (1H, *d*, *J* = 2.4, H-10c), 6.19 (1H, *t*, *J* = 2.1, H-12c), 6.43 (1H, *d*, *J* = 2.4, H-14c). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 133.4 (C-1a), 127.2 (C-2a/6a), 115.1 (C-3a/5a), 155.0 (C-4a), 85.0 (C-7a), 49.6 (C-8a), 146.2 (C-9a), 117.0 (C-10a), 158.0 (C-

14c).

11a), 100.0 (C-12a), 157.2 (C-13a), 103.0 (C-14a), 136.6 (C-1b), 128.7 (C-2b/6b), 114.4 (C-3b/5b), 157.3 (C-4b), 35.7 (C-7b), 50.9 (C-8b), 142.4 (C-9b), 118.2 (C-10b), 158.6 (C-11b), 95.0 (C-12b), 153.8 (C-13b), 121.5 (C-14b), 133.5 (C-1c), 129.0 (C-2c/6c), 114.6 (C-3c/5c), 157.2 (C-4c), 55.3 (C-7c), 66.6 (C-8c), 143.2 (C-9c), 107.5 (C-10c), 158.5 (C-11c), 100.1 (C-12c), 158.7 (C-13c), 107.3 (C-14c).

Stenophyllol B (9), obtained as a brown amorphous powder. MS *m/z*: 679 [MH<sup>-</sup>], m.p.: 255–257 °C. [α]<sub>D</sub><sup>20</sup>:  $-20^{\circ}$  (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 205, 228, 287 nm. IR (KBr) v<sub>max</sub> (cm<sup>-1</sup>): 3418 (OH), 1616, 1544, 1455 (C=C aromatic), and 831 (para-disubstituent). <sup>1</sup>H-NMR (acetone- $d_6$ , 300 MHz)  $\delta_H$  ppm: 6.88 (2H, d, J = 8.7, H-2a/6a), 6.77 (2H, *d*, *J* = 8.7, H-3a/5a), 5.84 (1H, *d*, *J* = 3.3, H-7a), 5.07 (1H, d, J = 3.3, H-8a), 6.31 (1H, d, J = 2.1, H-12a), 6.25 (1H, d, J = 2.1, H-14a), 7.20 (2H, d, J = 8.4, H-2b/6b), 6.66 (2H, *d*, *J* = 8.4, H-3b/5b), 4.73 (1H, *d*, *J* = 6.3, H-7b), 4.73 (1H, *d*, *J* = 6.3, H-8b), 6.79 (1H, *s*, H-14b), 7.29 (2H, d, J = 8.1, H-2c/6c), 6.68 (2H, d, J = 8.1, H-3c/5c), 5.35 (1H, *d*, *J* = 9.6 H-7c), 4.30 (1H, *dd*, *J* = 10.5, 8.4, H-8c), 6.07 (1H, m, H-12c), 6.07 (1H, m, H-14c). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 135.5 (C-1a), 128.2 (C-2a/6a), 116.9 (C-3a/5a), 158.7 (C-4a), 89.0 (C-7a), 53.5 (C-8a), 142.2 (C-9a), 124.5 (C-10a), 157.5 (C-11a), 102.4 (C-12a), 159.8 (C-13a), 107.8 (C-14a), 137.8 (C-1b), 130.9 (C-2b/6b), 116.8 (C-3b/5b), 157.1 (C-4b), 52.8 (C-7b), 57.4 (C-8b), 145.2 (C-9b), 121.4 (C-10b), 161.4 (C-11b), 96.8 (C-12b), 160.1 (C-13b), 109.3 (C-14b), 140.6 (C-1c), 130.8 (C-2c/6c), 116.8 (C-3c/5c), 157.2 (C-4c), 48.2 (C-7c), 54.5 (C-8c), 151.8 (C-9c), 124.4 (C-10c), 155.7 (C-11c), 100.1 (C-12c), 158.7 (C-13c), 107.3 (C-14c).

**Ampelopsin E (10)**, obtained as a reddish yellow, MS m/z: 679 [M<sup>+</sup>]. m.p.: 180–182 °C. [ $\alpha$ ]<sub>D</sub><sup>20</sup>: -94° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 230, 325 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3367 (OH), 2947 (C–H aliphatic), 1655, 1452 (C=C aromatic). <sup>1</sup>H-NMR (methanol- $d_4$ , 300 MHz)  $\delta_{\rm H}$  ppm: 7.27 (2H, d, J = 8.4, H-2a/6a), 6.84 (2H, d, J = 8.4, H-3a/5a), 5.45 (1H, d, J = 4.8, H-7a), 4.53 (1H, d, J = 4.8 H-8a), 6.26 (1H, d, J = 2.4, H-10a), 6.26 (1H, d, J = 2.4, H-10a), 6.26 (1H, d, J = 2.4, H-14a), 6.62 (2H, d, J = 8.0, H-2b/6b), 6.59 (2H, d, J = 8.0

8.4, H-3b/5b), 6.63 (1H, *d*, *J* = 16.5, H-7b), 6.59 (1H, *d*, *J* = 16.5, H-8b), 6.44 (1H, s, H-12b), 7.28 (2H, d, J = 8.5, H-2c/6c), 6.87 (2H, *d*, *J* = 8.5, H-3c/5c), 5.45 (1H, *d*, *J* = 5.4, H-7c), 4.56 (1H, *d*, *J* = 4.8, H-8c), 6.23 (1H, *d*, *J* = 2.4, H-10c), 6.23 (1H, *t*, *J* = 2.0, H-12c), 6.26 (1H, *d*, *J* = 2.0, H-14c). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 134.0 (C-1a), 128.6 (C-2a/6a), 116.5 (C-3a/5a), 158.4 (C-4a), 94.1 (C-7a), 55.6 (C-8a), 147.3 (C-9a), 107.0 (C-10a), 160.0 (C-11a), 102.14 (C-12a), 160.0 (C-13a), 102.14 (C-14a), 133.7 (C-1b), 127.9 (C-2b/6b), 115.9 (C-3b/5b), 158.3 (C-4b), 124.6 (C-7b), 131.8 (C-8b), 130.2 (C-9b), 120.1 (C-10b), 162.5 (C-11b), 91.3 (C-12b), 162.5 (C-13b), 120.1 (C-14b), 134.0 (C-1c), 128.6 (C-2c/6c), 116.5 (C-3c/5c), 158.4 (C-4c), 94.1 (C-7c), 55.6 (C-8c), 147.3 (C-9c), 107.0 (C-10c), 160.0 (C-11c), 102.1 (C-12c), 160.0 (C-13c), 107.0 (C-14c).

**Isohopeaphenol (11)**, obtained as a pale yellow. m.p.: 272–275 °C. [a]<sub>D</sub><sup>20</sup>: -396° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 230, 284 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3367 (OH), 2947 (C-H aliphatic), 1655, 1452 (C=C aromatic). <sup>1</sup>H-NMR (acetone- $d_4$ , 300 MHz)  $\delta_{\rm H}$  ppm: 7.57 (2H, d, J = 8.7, H-2a/6a), 7.01 (2H, *d*, *J* = 8.7, H-3a/5a), 5.45 (1H, *brd*, *J* = 9.9, H-7a), 5.45 (1H, *brd*, *J* = 9.9, H-8a), 7.85 (1H, *brs*, 11a-OH), 6.39 (1H, *d*, *J* = 8.7, H-12a), 8.15 (1H, *brs*, 13a-OH), 6.39 (1H, d, J = 8.7, H-12a), 8.15 (1H, brs, H-13a-OH), 6.29 (1H, *d*, *J* = 2.4, H-14a), 6.39 (2H, *d*, *J* = 8.7, H-2b/6b), 6.34 (2H, *d*, *J* = 8.7, H-3b/5b), 7.80 (1H, *brs*, H-4b-OH), 5.16 (1H, *d*, *J* = 2.1, H-7b), 3.48 (1H, *brs*, H-8b), 5.85 (1H, *d*, *J* = 2.1, H-12b), 7.80 (1H, *brs*, H-13b-OH), 5.53 (1H, *d*, J = 2.1, H-14b). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 132.9 (C-1a), 129.7 (C-2a/6a), 115.7 (C-3a/5a), 158.3 (C-4a-OH), 92.6 (C-7a), 52.9 (C-8a), 140.9 (C-9a), 117.2 (C-10a), 157.9 (C-11a), 105.6 (C-12a), 156.3 (C-13a-OH), 106.2 (C-14a), 136.6 (C-1b), 129.0 (C-2b/6b), 114.9 (C-3b/5b), 154.4 (C-4b-OH), 42.5 (C-7b), 51.6 (C-8b), 139.9 (C-9b), 147.2 (C-10b), 159.6 (C-11b), 94.3 (C-12b), 158.3 (C-13b-OH), 110.4 (C-14b).

**Hopeaphenol (12),** obtained as a pale yellow. m.p.: 272– 275 °C. [α]<sub>D</sub><sup>20</sup>: -396° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 230, 284 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3367 (OH), 2947 (C–H aliphatic), 1655, 1452 (C=C aromatic). <sup>1</sup>H-NMR (acetone-*d*<sub>4</sub>, 300 MHz)  $\delta_{\rm H}$  ppm: 7.15 (2H, *d*, *J* = 8.4, H-2a/6a), 6.80 (2H, *d*, *J* = 8.4, H-3a/5a), 5.77 (1H, *d*, *J* = 13.8,H-7a), 4.26 (1H, *d*, *J* = 12.3, H-8a), 6.56 (1H, *d*, *J* = 2.4, H-12a), 6.31 (1H, *d*, *J* = 2.4, H-14a), 6.31 (1H, *d*, *J* = 2.4, H-14a), 6.31 (1H, *d*, *J* = 2.4, H-14a), 6.93 (2H, *d*, *J* = 8.4, H-2b/6b), 6.58 (2H, *d*, *J* = 8.4, H-3b/5b), 5.80 (1H, *s*, H-7b), 3.95 (1H, *brs*, H-8b), 5.74 (1H, *d*, *J* = 2.1, H-12b), 5.16 (1H, *d*, *J* = 2.1, H-14b). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 128.3 (C-1a), 129.3 (C-2a/6a), 114.9 (C-3a/5a), 157.7 (C-4a), 157.7 (C-7a), 87.3 (C-8a), 48.9 (C-9a), 141.6 (C-10a), 120.3 (C-11a), 101.6 (C-12a), 156.3 (C-13a), 105.6 (C-14a), 134.3 (C-1b), 128.4 (C-2b/6b), 114.1 (C-3b/5b), 154.7 (C-4b), 40.1 (C-7b), 51.6 (C-8b), 139.6 (C-9b), 117.9 (C-10b), 158.3 (C-11b), 94.3 (C-12b), 156.3 (C-13b), 110.3 (C-14b).

Vaticanol B (13), obtained as a brown amorphous powder. MS *m/z*: 905 [MH<sup>-</sup>]. m.p.: 205–207 °C. [a]<sub>D</sub><sup>20</sup>:  $-40^{\circ}$  (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 230, 284 nm. IR (KBr) v<sub>max</sub> (cm<sup>-1</sup>): 3367 (OH), 2947 (C-H aliphatic), 1655, 1452 (C=C aromatic). <sup>1</sup>H-NMR (methanol- $d_4$ , 500 MHz)  $\delta_{\rm H}$  ppm: 7.18 (2H, *d*, *J* = 8.5, H-2a/6a), 6.78 (2H, d, J = 8.5, H-3a/5a), 5.72 (1H, d, J = 12.0, H-7a), 4.33(1H, *d*, *J*= 12.0, H-8a), 6.18 (1H, *d*, *J* = 2.0, H-12a), 6.05 (1H, *s*, H-10a), 7.13 (2H, *d*, *J* = 8.5, H-2b/6b), 6.68 (2H, *d*, *J* = 8.5, H-3b/5b), 5.28 (1H, *d*, *J* = 5.5, H-7b), 3.15 (1H, *d*, *J* = 12.5, H-8b), 5.98 (1H, *s*, H-12b), 6.45 (2H, *d*, *J* = 8.5, H-2c/6c), 6.49 (2H, *d*, *J* = 8.5, H-3c/5c), 4.08 (1H, *t*, J = 11.5, H-7c), 4.42 (1H, d, J = 10.5, H-8c), 6.19 (1H, s, H-12c), 6.44 (1H, *d*, *J* = 1.5, H-14c), 7.14 (2H, *d*, *J* = 8.5, H-2d/6d), 6.75 (2H, *d*, *J* = 8.5, H-3d/5d), 5.28 (1H, *d*, *J* = 5.5, H-7d), 5.99 (2H, *d*, *J* = 2.5, H-10d/14d), 6.20 (1H, *d*, J = 2.0, H-12d).<sup>13</sup>C-NMR (125 MHz)  $\delta_{\text{C}}$  ppm: 129.7 (C-1a), 130.9 (C-2a/6a), 114.9 (C-3a/5a), 157.9 (C-4a), 89.6 (C-7a), 49.3 (C-8a), 141.3 (C-9a), 124.3 (C-10a), 154.4 (C-11a), 100.5 (C-12a), 156.9 (C-13a), 100.9 (C-14a), 132.9 (C-1b), 129.4 (C-2b/6b), 113.8 (C-3b/5b), 154.7 (C-4b), 35.8 (C-7b), 51.9 (C-8b), 147.2 (C-10b), 113.8 (C-11b), 154.7 (C-12b), 35.8 (C-13b), 51.9 (C-8b), 147.2 (C-9b), 113.8 (C-10b), 158.3 (C-11b), 94.3 (C-12b), 154.0 (C-13b), 121.6 (C-14b), 130.9 (C-1c), 129.0 (C-2c/6c), 114.2 (C-3c/5c), 155.3 (C-4c), 57.4 (C-7c), 49.3 (C-8c), 140.7 (C-9c), 122.5 (C-10c), 160.8 (C-11c), 94.2 (C-12c), 159.6 (C-13c), 104.4 (C-14c), 133.6 (C-1d), 127.3 (C-2d/6d), 114.9 (C-3d/5d), 157.4 (C-4d), 89.6 (C-7d), 56.5 (C-8d), 142.5 (C-9d), 106.1 (C-10d/14d), 160.4 (C-11d/13d), 100.5 (C-12d).

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Diptoindonesin E (14), obtained as a white amorphous powder. MS *m/z*: 903 [MH<sup>-</sup>]. m.p.: 233–235 °C. [α]<sub>D</sub><sup>20</sup>:  $-95^{\circ}$  (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 205, 228, 325 nm. IR (KBr) v<sub>max</sub> (cm<sup>-1</sup>): 3401 (OH), 2922 (C–H aliphatic), 1655, 1452 (C=C aromatic). <sup>1</sup>H-NMR (methanol- $d_4$ , 300 MHz)  $\delta_{\rm H}$  ppm: 7.27 (2H, d, J = 8.7, H-2a/6a), 6.88 (2H, *d*, *J* = 8.7, H-3a/5a), 5.47 (1H, *d*, *J* = 12.0, H-7a), 4.69 (1H, *d*, *J*=3.9, H-8a), 6.26 (2H, *d*, *J* = 2.1, H-10a/14a), 6.19 (1H, *t*, *J* = 2.1, H-12a), 7.70 (2H, *d*, *J* = 2.1, H-2b/6b), 6.74 (2H, *d*, *J* = 8.5, H-3b/5b), 6.78 (1H, *d*, *J* = 5.5, H-7b), 6.70 (1H, *d*, *J* = 16.5, H-8b), 6.42 (1H, *s*, H-12b), 6.46 (2H, *d*, *J* = 8.7, H-2c/6c), 6.52 (2H, *d*, *J* = 8.7, H-3c/5c), 5.04 (1H, *d*, *J* = 1.9, H-7c), 4.76 (1H, *d*, *J* = 1.9, H-8c), 6.23 (1H, *d*, *J* = 2.2, H-12c), 5.99 (1H, *d*, *J* = 2.1, H-14c), 7.50 (1H, *d*, *J* = 2.4, H-2d), 6.89 (1H, *d*, *J* = 8.7, H-5d), 7.23 (*dd*, *J* = 9.0, 2.1, H-6d), 5.18 (1H, *d*, *J* = 1.5, H-7d), 4.79 (1H, *d*, *J* = 1.6, H-8d), 5.95 (2H, *brd*, *J* = 2.1, H-10d/14d), 6.29 (1H, *t*, *J* = 2.1, H-12d). <sup>13</sup>C-NMR (75 MHz) δ<sub>C</sub> ppm: 133.3 (C-1a), 126.8 (C-2a/6a), 116.2 (C-3a/5a), 158.9 (C-4a), 93.6 (C-7a), 57.1 (C-8a), 141.7 (C-9a), 106.0 (C-10a/14a), 159.4 (C-11a/13a), 101.4 (C-12a), 131.5 (C-1b), 130.8 (C-2b/6b), 126.2 (C-3b), 153.8 (C-4b), 116.9 (C-5b), 128.8 (C-6b), 131.1 (C-7b), 126.9 (C-8b), 131.1 (C-9b), 115.9 (C-10b), 162.5 (C-11b), 91.6 (C-12b), 161.9 (C-13b), 122.0 (C-14b), 132.9 (C-1c), 126.9 (C-2c/6c), 115.6 (C-3c/5c), 157.4 (C-4c), 90.6 (C-7c), 51.4 (C-8c), 145.6 (C-9c), 118.9 (C-10c), 162.7 (C-11c), 95.9 (C-12c), 161.8 (C-13c), 107.2 (C-14c), 135.6 (C-1d), 131.9 (C-2d/6d), 128.4 (C-3d), 156.8 (C-4d), 115.5 (C-5d), 91.5 (C-7d), 55.2 (C-8d), 147.2 (C-9d), 106.1 (C-10d/14d), 161.8 (C-11d/13d), 102.1 (C-12d).

Hemsleyanol D (15), obtained as a brownish-yellow solid. MS *m/z*: 905 [MH<sup>-</sup>]. m.p.: 280–282 °C.  $[\alpha]_D^{20}$ : +29° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 203, 230, 284 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3400 (OH), 2927 (C–H aliphatic), 1614, 1512 (C=C aromatic). <sup>1</sup>H-NMR (acetone-*d*<sub>6</sub>, 300 MHz)  $\delta_H$  ppm: 7.22 (2H, *d*, *J* = 8.7, H-2a/6a), 6.78 (2H, *d*, *J* = 8.5, H-3a/5a), 5.77 (1H, *d*, *J* = 11.7, H-7a), 4.41 (1H, *d*, *J* = 11.7, H-8a), 6.36 (1H, *d*, *J* = 2.4, H-12a), 6.12 (1H, *d*, *J* = H-10a), 6.94 (2H, *d*, *J* = 8.7, H-2b/6b), 6.48 (2H, *d*, *J* = 8.7, H-3b/5b), 5.29 (1H, *d*, *J* = 3.4, H-7b), 3.38 (1H, *d*, *J* = 10.9, H-8b), 6.02 (1H, *s*, H-12b), 6.72 (2H, *d*, *J* = 8.7, H-2c/6c), 6.52 (2H, *d*, *J* = 8.7, H-3c/5c), 4.55 (1H, *d*, *J* = 10.2, H-7c),

3.89 (1H, *dd*, *J* = 11.7, 10.8, H-8c), 6.23 (1H, *d*, *J* = 2.0, H-12c), 6.79 (1H, s, H-14c), 7.06 (2H, d, J = 8.4, H-2d/6d), 6.82 (2H, d, J = 8.4, H-3d/5d), 4.92 (1H, d, J = 1.5, H-7d), 3.50 (1H, brs, H-8d), 5.34 (2H, brs, H-10d/14d), 6.07 (1H, t, J = 2.1 H-12d). <sup>13</sup>C-NMR (75 MHz) δ<sub>C</sub> ppm: 132.5 (C-1a), 129.9 (C-2a/6a), 115.3 (C-3a/5a), 157.7 (C-4a), 89.6 (C-7a), 48.0 (C-8a), 140.7 (C-9a), 124.0 (C-10a), 154.9 (C-11a), 100.6 (C-12a), 155.9 (C-13a), 104.9 (C-14a), 133.9 (C-1b), 129.3 (C-2b/6b), 115.3 (C-3b/5b), 157.1 (C-4b), 36.2 (C-7b), 56.5 (C-8b), 142.1 (C-10b), 114.9 (C-11b), 158.4 (C-12b), 153.8 (C-13b), 120.4 (C-14b), 132.5 (C-1c), 128.4 (C-2c/6c), 114.7 (C-3c/5c), 155.8 (C-4c), 53.1 (C-7c), 57.4 (C-8c), 140.1 (C-9c), 94.8 (C-10c), 162.2 (C-11c), 116.3 (C-12c), 159.5 (C-13c), 104.9 (C-14c), 136.4 (C-1d), 127.1 (C-2d/6d), 115.3 (C-3d/5d), 154.8 (C-4d), 93.1 (C-7d), 60.1 (C-8d), 147.1 (C-9d), 105.5 (C-10d/14d), 158.1 (C-11d/13d), 101.2 (C-12d).

**Bergenin (16)**, obtained as a white crystal. MS *m/z*: 327 [MH<sup>-</sup>]. m.p.: 244–246 °C.  $[\alpha]_D^{20}$ : -30° (c 0.1 MeOH). UV (MeOH)  $\lambda_{max}$ : 307, 274 nm. IR (KBr)  $v_{max}$  (cm<sup>-1</sup>): 3420 (OH), 2927 (C–H aliphatic), 1614, 1512 (C=C aromatic), 1703 (C=O), 2949 (C-H). <sup>1</sup>H-NMR (acetone-*d*<sub>6</sub>, 500 MHz)  $\delta_H$  ppm: 4.00 (1H, *dd*, *J* = 10.0, 4.0, H-2), 3.45 (1H, *t*, *J* = 9.0, H-3), 3.80 (1H, *t*, *J* = 9.0, H-4), 3.70 (1H, *dd*, *J* =9.5, 7.0, H-4A), 7.08 (1H, *s*, H=7), 4.94 (1H, *d*, *J* = 10.8, H-10B), 3.65 (1H, *m*, H-11A), 4.10 (1H, *dd*, *J* = 9.5, 4.0, H-11B), 3.89 (1H, *s*, OMe).<sup>13</sup>C-NMR (125 MHz)  $\delta_C$ ppm: 81.5 (C-2), 71.9 (C-3), 75.7 (C-4), 83.0 (C-4A), 165.8 (C-6), 119.52 (C-6A), 111.1 (C-7), 152.42 (C-8), 142.3 (C-9), 149.5 (C-10), 117.3 (C-10A), 74.32 (C-10B), 62.72 (C-11), 60.92 (C-Ome).

**Scopoletin (17)**, obtained as a white powder. m.p.: 171– 175 °C. UV (MeOH)  $\lambda_{max}$ : 256, 342 nm. IR (KBr)  $\nu_{max}$ (cm<sup>-1</sup>): 3536 (OH), 2927 (C–H aliphatic), 1700, 1635 (C=O conjugated), 1616, 1562, 1461 (C=C aromatic), 1288, 1140 (C-O oxyaryl). <sup>1</sup>H-NMR (methanol- $d_4$ , 300 MHz)  $\delta_{\rm H}$  ppm: 6.20 (1H, d, J = 9.3, H-3), 7.84 (1H, d, J = 9.3, H-4), 7.12 (1H, *s*, H-5), 6.78 (1H, *s*, H-8), 3.92 (3H, *s*, OCH<sub>3</sub>). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 161.4 (C-2), 113.3 (C-3), 144.7 (C-4), 112.1 (C-4a), 109.9 (C-5), 146.0 (C-6), 151.9 (C-7), 103.8 (C-8), 151.2 (C-8a), 56.7 (C-OMe). **4-O'-methylgallocatechin** (18), obtained as an amorphous pale-yellow needle solid. MS *m/z*: 319 [MH<sup>-</sup>]. m.p.: 156–157 °C (dec.). UV (MeOH)  $\lambda_{max}$ : 239, 274 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3518, 1602 (C=C), and 1461. <sup>1</sup>H-NMR (methanol, 300 MHz)  $\delta_{\rm H}$  ppm: 4.59 (1H, overlapped, H-2), 3.99 (1H, *m*, H-3), 2.84 (1H, *dd*, *J* = 16.2, 5.1, H-4α), 2.56 (1H, *dd*, *J* = 16.2, 7.8, H-4β), 5.95 (1H, *d*, *J* = 2.1, H-6), 5.86 (1H, *d*, *J* = 2.1, H-8), 6.42 (1H, *s*, H-2'), 6.42 (1H, *s*, H-6'), 3.8 (3H, *s*, OMe). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 81.2 (C-1), 67.4 (C-2), 26.8 (C-3), 99.3 (C-4α/4β), 95.0 (C-5), 156.4 (C-6), 94.2 (C-1), 155.3 (C-1), 135.3 (C-1), 106.0 (C-1), 150.2 (C-1), 135.2 (C-1), 150.2 (C-1), 106.0 (C-1), 59.4 (C-1).

**4-O'-methylepigallocatechin** (19), obtained as an amorphous pale-yellow needle solid. MS *m/z*: 639 [MH<sup>-</sup>]. m.p.: 156–157 °C (dec.). UV (MeOH)  $\lambda_{max}$ : 239, 274 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3518, 1602 (C=C), and 1461. <sup>1</sup>H-NMR (methanol, 500 MHz)  $\delta_{\rm H}$  ppm: 4.82 (1H, *s*, H-2), 4.20 (1H, *m*, H-3), 2.73 (1H, *dd*, *J* = 16.5, 3.0, H-4α), 2.86 (1H, *dd*, *J* = 16.5, 3.0, H-4β), 6.01 (1H, *d*, *J* = 2.0, H-6), 5.91 (1H, *d*, *J* = 2.0, H-8), 6.58 (1H, *s*, H-2'/6'), 3.78 (3H, *s*, OMe). <sup>13</sup>C-NMR (125 MHz)  $\delta_{\rm C}$  ppm: 9.2 (C-1), 66.8 (C-2), 28.3 (C-4α/4β), 99.7 (C-4a), 156.6 (C-6), 96.1 (C-6), 95.3 (C-8), 157.5 (C-8a), 136.2 (C-1'), 107.0 (C-2'/6'), 150.8 (C-3'/5'), 136.4 (C-4'), 60.5 (OMe).

4-methoxy-epigallocatechin-3-0-(4-methyl) gallate (20), obtained as an amorphous light yellow solid. MS *m/z*: 485, 319, 274 [MH<sup>-</sup>]. m.p.: 192–195 °C. UV (MeOH)  $\lambda_{max}$ : 224, 284 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3423 (OH), 2935 (C–H), 1054 (C–O). <sup>1</sup>H-NMR (methanol- $d_4$ , 300 MHz)  $\delta_{\rm H}$ ppm: 5.1 (1H, s, H-2), 5.5 (1H, m, H-3), 2.98 (1H, dd, J = 17.1, 4.5, H-4 $\alpha$ ), 3.06 (1H, dd, J = 17.1, 4.5, H-4 $\beta$ ), 6.03 (1H, *d*, *J* = 2.1, H-6), 5.99 (1H, *d*, *J* = 2.4, H-8), 6.56 (1H, s, H-2'/6'), 3.76 (3H, s, OMe), 7.16 (1H, s, H-2'/6", 7.16 (1H, s, H-6"), 3.87 (1H, s, OMe). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$ ppm: 76.8 (C-2), 69.4 (C-2), 26.7 (C-4α/4β), 97.9 (C-4a), 155.6 (C-5/7), 95.3 (C-6), 94.8 (C-8a), 151.0 (C-1'), 127.6 (C-2'/6'), 105.6 (C-1), 147.4 (C-3'/5'), 134.5 (C-4'), 59.5 (OMe), 120.0 (C-1"), 106.2 (C-2"), 147.4 (C-3"), 143.0 (C-4"), 144.4 (C-5"), 106.2 (C-6"), 166.2 (CO), 55.4 (OMe). β-sitosterol (21), obtained as a whitish solid. m.p: 287-295 °C. UV (MeOH)  $\lambda_{max}$ : 210 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3423 (OH), 2935 (C-H), 1054 (C-O). <sup>1</sup>H-NMR (methanol- $d_4$ , 300 MHz)  $\delta_{\rm H}$  ppm: 3.53 (1H, tdd, J = 4.5, 4.2, 3.8, H-2), 5.36 (1H, t, J = 6.4, H-5), 0.93 (1H, d, J = 6.5, H-19), 0.84 (1H, t, J = 7.2, H-24), 0.83 (1H, d, J = 6.4, H-26), 0.81 (1H, d, J = 6.4, H-27), 0.68 (1H, s, H-28), 1.01 (1H, s, H-29). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 37.2 (C1), 31.6 (C2), 71.8 (C3), 42.3 (C4), 140.8 (C5), 121.7 (C6), 31.9 (C7), 31.9 (C-8), 50.1 (C-9), 36.5 (C-10), 21.7 (C-11), 39.8 (C-12), 42.3 (C-13), 56.8 (C-14), 24.3 (C-15), 28.2 (C-16), 56.0 (C-17), 11.9 (C-18), 19.4 (C-19), 36.1 (C-20), 19.8 (C-21), 33.9 (C-22), 26.1 (C-23), 45.8 (C-24), 29.1 (C-25), 19.8 (C-26), 19.0 (C-27), 23.1 (C-28), 12.0 (C-29).

β-sitosterol-3-O-β-D-glucoside (22), obtained as a whitish solid. MS m/z: 545 [M-H<sub>2</sub>O]. m.p.: 287-295 °C. UV (MeOH)  $\lambda_{max}$ : 210 nm. IR (KBr)  $\nu_{max}$  (cm<sup>-1</sup>): 3423 (OH), 2935 (C-H), 1054 (C-O). <sup>1</sup>H-NMR (methanol-d<sub>4</sub>, 300 MHz) δ<sub>H</sub> ppm: 3.87 (1H, *m*, H-3), 5.40 (1H, *m*, H-6), 4.30 (1H, *d*, *J* = 7.7, H-1'), 3.1 (4H, *m*, H-2', H-3', H-4', H-5'), 4.90 (1H, dd, J = 10.6, 2.7, H-6a'), 4.50 (1H, t, J = 5/7, H-6b'). <sup>13</sup>C-NMR (75 MHz)  $\delta_{\rm C}$  ppm: 36.4 (C-1), 33.8 (C-2), 77.2 (C-3), 38.7 (C-4), 140.9 (C-5), 121.7 (C-6), 31.8 (C-7), 31.9 (C-8), 50.1 (C-9), 36.7 (C-10), 20.2 (C-11), 37.2 (C-12), 42.3 (C-13), 56.7 (C-14), 58.1 (C-15), 28.3 (C-16), 55.9 (C-17), 12.2 (C-18), 19.6 (C-19), 35.6 (C-20), 19.1 (C-21), 33.5 (C-22), 29.1 (C-23), 45.6 (C-24), 29.8 (C-25), 19.4 (26), 23.2 (C-27), 24.7 (C-28), 12.1 (C-29), 101.2 (C-1'), 73.9 (C-2'), 77.2 (C-3'), 70.5 (C-4'), 77.4 (C-5'), 61.5 (C-6').

It is interesting to note that the polymerization of oligomeric resveratrol is significantly larger and more diversified in the tribe of Dipterocarpeae as compared to the tribe of Shoreae [42]. In Shoreae, this study discovered an inclination in the production of tetramer from monomer whereas Dipterocarpeae showed more variations from monomer up to octamer (Table 1). Matsuda et al. [43] indicated that the type of biogenetic of initial oligomerization of oligomeric resveratrol in Dipterocarpaceae and other families that produce oligomer resveratrol are different from each other. This supports the fact that a similar type of compound comes from the plant of the same tribe and family.

Table 2 indicates resveratrol oligomers isolated inthe Dipterocarpus genus. Currently, the polymerization

of resveratrol in the genus *Dipterocarpus* occurred from its monomer to tetramer. Dimer and tetramer resveratrol are the most abundant compounds. This is supported by a previous study [3] which disclosed that resveratrol tetramers and dimers are the principal oligomers isolated from *Dipterocarpus*.

Table 3 tabulated the distribution of oligomer resveratrol isolated in *Dipterocarpus* study from the tribe of Dipterocarpeae. Dimer resveratrol, (-)- $\varepsilon$ -viniferin (2), have been isolated in all *Dipterocarpus*. The presence of  $\varepsilon$ viniferin (2) has no chemotaxonomic significance as it is regarded as the general precursor for oligostilbenoids. Laevifonol (3) which is a unique oligostilbenoid formed from a condensation of (-)- $\varepsilon$ -viniferin (2) and ascorbic acid highlights the relationship between *Dipterocarpus* and *Vatica* since previous research stated that these metabolites can only be found in *Vatica umbonata* and *Vatica odorata* in Dipterocarpaceae. Another dimer resveratrol, ampelopsin F (4) with the skeleton bicyclo[3.2.1]octane found in *D. grandiflorus*, *U. borneensis*, *V. mangachapoi*, and *C. melanoxylin*, indicated that these metabolites have a significant relationship with those genera. In addition, ampelopsin A (5) with the skeleton of benzofuran-cycloheptane has

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Species	а	b	С	d	e	f	g	h	References	
D. grandiflorus	-	5	1	6	-	-	-	-	[5]	
D. retusus	-	1	2	-	-	-	-	-	[4]	
D. hasseltii	-	2	1	3	-	-	-	-	[4]	
D. alatus	-	-	-	1	-	-	-	-	[11]	
D. intricatus	-	1	-	1	-	-	-	-	[8]	
D. semivestitus	-	-	1	2	-	-	-	-	[9-10]	
D. verrucosus	-	2	1	4	-	-	-	-	[6], present study	
D. crinitus	1	2	2		-	-	-	-	[7], present study	
D. cornutus	-	3	2	1	-	-	-	-	present study	
V. rassak	2	1	3	4	-	3	1	-	[44-47]	
V. pauciflora	2	8	10	8	-	-	1	-	[48-50]	
V. odorata	-	1	2	2	-	-	-	-	[51]	
V. umbonata	-	3	2	2	-	-	-	-	[52]	
V. diospyroides	-	-	-	2	-	-	-	-	[53]	
V. albiramis	1	7	-	6	-	1	-	1	[54]	
V. affinis	-	1	-	1	-	-	-	-	[55]	
V. oblongifolia	-	-	-	3	-	-	-	-	[56]	
V. mangachapoi	-	10	5	6	-	-	-	-	[57]	
V. chinensis	-	-	-	2	-	-	-	-	[58-59]	
V. lowii	1	-	-	-	-	-	-	-	[50]	
V. bantamensis	-	-	-	1	-	-	-	-	[60]	
U. borneensis	5	6	-	18	4	1	-	1	[61]	
A. laevis	-	-	-	1	-	-	-	-	[62]	
A. marginata	-	2	-	3	-	-	-	-	[63]	
A. thurifera	-	-	-	3	-	-	-	-	[64]	
S. canaliculatus	-	1	2	-	-	-	-	-	[65]	
V. indica	1	2	-	11	-	-	-	1	[66]	
V. copallifera	-	-	3	-	-	-	-	-	[67,68]	
C. lanceolatum	1	-	2	1	-	-	-	-	[69]	
C. melanoxvlon	-	5	4	_	-	-	-	-	[70]	

Table 1. Distribution of oligomer resveratrols in Dipterocarpeae tribe

\*a = monomer, b = dimer, c = trimer d = tetramer, e = pentamer, f = hexamer, g = heptamer, h = octamer

Compounds	Type	DG	DH	DR	DA	DI	DS	DV	DC	DCI
Resveratrol	monomer	20	211	211	211	21	20	21	20	<u></u> √
ε-Viniferin	dimer									Ń
Ampelopsin A	dimer	Ń	•	,		,		,	•	Ń
Laevifonol	dimer	,								•
Shorealactone	dimer		,					,	•	
Ampelopsin F	dimer	Ń								
Miyabenol C	dimer	Ń							•	
a-Viniferin	trimer	J								
Vaticanol A	trimer	,	,	J			•	,		V
Stepophyllol B	trimer			•						•
Davidiol A	trimer								J	
Ampelopsin F	trimer	N							v	
Isobonomonol	totromor	v						2		
Isonopeapenoi	tetramer	al	2				2	N		
Hopeapenol	tetramer	N	N	.1			V	N		
Vaticanol B	tetramer	γ	N	N				N		
Diptoindonesin E	tetramer									
Vaticanol C	tetramer									
Hemsleyanol D	tetramer						$\checkmark$			
Grandiphenol A	tetramer									
Grandiphenol B	tetramer	$\checkmark$								
Vaticaffinol	tetramer				$\checkmark$					

Table 2. Oligomer resveratrol isolated from genus Dipterocarpus

\*DG = D. grandiflorus, DH = D. hasseltii, DR = D. retusus, DA=D. alatus, DI = D. intricatus, DS = D. semivestitus, DV = D. vertucosus, DC = D. crinitus, DCJ = D. cornutus

<b>Table 3.</b> The distribution of oligomer	resveratrol isolated in Dipterocar	<i>pus</i> study in tribe Dipterocarpaceae

Isolated compounds					Dipteroc	arpaceae			
	А	В	С	D	Е	F	G	Н	Ι
Monomer									
(+)Resveratrol	$\checkmark$		$\checkmark$						
Dimers									
(-)-ε-Viniferin	$\checkmark$	$\checkmark$							$\checkmark$
(-)-Ampelopsin A	$\checkmark$				$\checkmark$				
(-)-Laevifonol	$\checkmark$					$\checkmark$			$\checkmark$
(-)-Ampelopsin F		$\checkmark$							
(+)-Ampelopsin F	$\checkmark$		$\checkmark$			$\checkmark$			
Trimers									
(+)-α-Viniferin									$\checkmark$
(-)-Vaticanol A	$\checkmark$					$\checkmark$			
(-)-Stenophyllol B	$\checkmark$								
(-)-Davidiol A	$\checkmark$	$\checkmark$							
(-)-Ampelopsin E									$\checkmark$
Tetramers									
(-)-Isohopeaphenol					$\checkmark$				
(-)-Hopeaphenol	$\checkmark$								$\checkmark$
(-)-Vaticanol B	$\checkmark$		$\checkmark$		$\checkmark$				
(+)-Diptoindonesin E		$\checkmark$							
Hemsleyanol D	$\checkmark$								
Total	11	12	3	3	3	3	0	0	5

\*A-Vatica, B-Dipterocarpus, C-Upuna, D-Anisoptera, E-Vateria, F-Cotylelobium, G-Vateriopsis, H-Stemonoporus, I-Dryobalanops

been previously reported from *D. grandiflorus*, *Anisoptera marginata*, *V. albiramis*, *V. mangachapoi*, and *Vateria indica*. The occurrence of  $\alpha$ -viniferin (6) as a trimer resveratrol in most of *Dipterocarpus* can be quantified, whereby this compound acts as a chemical marker for genus *Dipterocarpus* since these metabolites are not detected in other genera in the subtribe Dipterocarpeae. However, this metabolite is not found in *D. cornutus*. The occurrence of vaticanol A (7), which is also a trimer resveratrol indicated the significant relationship between *Dipterocarpus* and other previously isolated genera from *V. rassak*, followed by *V. pauciflora*, *D. retusus*, *Cotylelobium melanoxylin*, and *V. mangachapoi*.

The significant findings in this study are the occurrence of resveratrol (1), davidiol A (8), stenophyllol B (9), ampelopsin E (10), and isohopeaphenol (12), which for the first time reported in *Dipterocarpus*. Resveratrol (1) acts as a monomer isolated from *D. crinitus* indicating another strong evidence that further correlates the relationship between *Dipterocarpus* and *Vatica*. The previous study only discussed the occurrence in *V. rassak* and *U. borneensis*.

This finding supports the theory of polymerization of oligomer resveratrol which suggests that the starting material is resveratrol, which acts as a precursor compound. This is the new in contrast to the previous studies (D. grandiflorus, hasseltii, and retusus), biogenetically, that suggested the role of  $\varepsilon$ -viniferin as a precursor. The presence of davidiol A (8) for the first time in Dipterocarpus, as well as its occurrence in V. mangachapoi portrayed diversifying attributes of trimer resveratrol in Dipterocarpus. Meanwhile, the isolation of stenophyllol B (9) was also reported for the first time in Dipterocarpus, resulting in another strong and significant relationship between Dipterocarpus and Vatica. Based on previous research, this metabolite can only be found in V. umbonata and V. pauciflora. The presence of isohopeaphenol (11), tetramer oligostilbenoid, is the second occurrence in Dipterocarpaceae after V. indica.

Diptoindonesin E (14), tetramer resveratrol, gives the second isolation after *D. hasseltii*, and until now, the compound has not yet been isolated in any genus of Dipterocarpaceae as well as in any family. The  $^{13}$ C-NMR result indicated that it is consistent with the structure of amurensin J, which was isolated from Vitis amurensis [71] from the Vitaceae family. However, the occurrence of the bridge at C-3b and C-3d of diptoindonesin E (14) shows that both compounds are different. Despite the small difference in the structure, the result provided other attributes in terms of the affinity of Dipterocarpus with Dryobalanops. This finding was supported by the isolation of flexuosol A for the first time from Dryobalanops lanceolata [72]. Amurensin J is a stereoisomer of flexuosol A. The isolation of diptoindonesin E (14) consequently produced a close structure with both compounds of amurensin J and flexuosol A. This is a convincing result to support the chemotaxonomy attributes of Dipterocarpus in Dipterocarpeae. The phylogenetic placement of Dipterocarpus and Dryobalanops remains unresolved. For that reason, this is an alarming call for further study to facilitate and enhance the holistic comprehension of phylogenetic and generic limitations of Dipterocarpaceae.

Moreover, the first isolation of ampelopsin E (10) in *Dipterocarpus* resulted in a strong correlation between *Dipterocarpus* and *Dryobalanops*. Previous research only involves the isolation of ampelopsin E from *Dryobalanops aromatica* [73-74]. This reveals the strong chemotaxonomy correlation between the species in Dipterocarpaceae. Stereoisomers of isohopeahenol (12), hopeaphenol (13), were isolated and these metabolites are classified as a chemical marker in Dipterocarpaceaea [6], which was previously found in *D. grandiflorus*, *D. hasseltii*, *Vateria indica*, *Anisoptera marginata*, *V. umbonate*, and *V. albiramis*.

The presence of vaticanol B (13), which is common and increasingly isolated in Dipterocarpaceae family shows the chemotaxonomically-correlated relationship among V. rassak, Vateria indica, V. pauciflora, A. marginata, V. umbonata, V. pauciflora, D. grandiflorius, U. borneensis, V. indica, V. albiramis, and D. hasseltii. The presence of hemsleyanol D (15), tetramer resveratrol which was isolated in D. cornutus also attained the relationship of chemotaxonomy characteristics between Dipterocarpus and Vatica. Previously, this metabolite was only isolated in D. grandiflorus and from the other genus, namely V. pauciflora, and V. mangachapoi.

This study also discovered the presence of several major non-oligomeric resveratrol. Bergenin (15) and scopoletin (16) are both coumarins and can be classified as chemical markers in Dipterocarpaceae, which can be found abundantly. The occurrence of terpenes,  $\beta$ -sitosterol (17) and  $\beta$ -sitosterol glucoside (18) are also common in Dipterocarpaceae and most plant kingdoms. However, the presence of two flavonoids, 4-*O*-methyl gallocatechin (18) and 4-*O*-methyl epigallocatechin (19) are only reported in the family other than Dipterocarpaceae and the 4-methoxy-epigallocatechin-3-*O*-(3-methyl) gallate (20), is firstly reported on the occurrence in the plant kingdom.

The chemotaxonomic classification significantly showed that *Dipterocarpus* shares many isolated compounds similar to *Vatica*. Therefore, these data suggested that the significant chemotaxonomic relationship between *Dipterocarpus* and *Vatica* are closely related to each other. It is forecasted that *Dipterocarpus* will be inclined to produce octamer resveratrol, as well as *Vatica*. In addition, it also indicated in this research that the relationship of *Dipteocarpus* was supported by another report [1] in terms of its phylogenetic classification, which consists of *Dipterocarpus, Anisoptera, Cotylelobium, Stemonoporus, Upuna, Vateria, Vateriopsis*, and *Vatica*.

# CONCLUSION

This research found that Dipterocarpaceae comprise oligomeric and non-oligomeric compounds that can be isolated via different chromatographic techniques. This is an imperative discovery as it will help to facilitate the investigation to quantify the relationship among the species and genera of Dipterocarpaceae. Additionally, the findings suggested that *Dipterocarpus* and *Vatica* are closely connected in terms of chemotaxonomic relationships.

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# CONFLICT OF INTEREST

This research does not have a conflict of interest.

# AUTHOR CONTRIBUTIONS

Wan Zuraida Wan Mohd Zain, Liliwirianis Nawi, Aisyah Salihah Kamarozaman, Noorazlina Adnan and Siti Zakirah Azahar composed the original draft, refined the literature review, and edited the writing format. Norizan Ahmat, Che Puteh Othman and Yoshiaki Takaya designed the methodology. All authors collectively approved the final manuscript.

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