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The Chemistry and Biological Activities of Secondary Metabolites from the Dipterocarpaceae Family: A Review

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The Dipterocarpaceae family, one of the largest plant families, is crucial for Southeast Asia's biodiversity and holds a significant economic value due to its timber. Rich in secondary metabolites like oligostilbenoids, this family displays unique structural variations that facilitate taxonomic classification and genetic understanding. Oligostilbenoids and related compounds demonstrate diverse biological activities, such as anticancer, antimicrobial, and anti-inflammatory effects, suggesting their potential as novel therapeutic agents. This review explores the chemical diversity of secondary metabolites in the Dipterocarpaceae family, highlighting their structural diversity, biogenesis, biological properties, therapeutic potentials, and the challenges related to their bioactivity and applications. It also emphasizes the need for further research to fully understand the ecological roles and pharmacological potential of these compounds.

Keywords: Dipterocarpaceae; oligostilbenoids; terpenes; flavonoid; biogenesis; bioactivity

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Dipterocarpaceae (dipterocarps) is a family of large trees commonly found in the tropical rainforests of Southeast Asia, particularly in Borneo and Peninsular Malaysia. These trees typically exceed 50 meters (150 feet) in height and 1 meter in diameter. They spread widely, forming dense canopies above the forest floor. A dipterocarp forest is rather dark due to its thick, stable canopy. Members of this family play a vital role in the economies of many Southeast Asian countries, given the significance of dipterocarp timber products in the international tropical timber market [1]. Dipterocarpaceae can typically be divided into three subfamilies: Dipterocarpoideae, Monotoideae, and Pakaraimoideae, which consist of 17 genera and approximately 511 species. The distribution of dipterocarp species spans the tropical regions of Asia, Africa, and South America [2].

The chemical constituents of Dipterocarpaceae have been extensively studied [3, 4], yet their overall chemical structure and role remains less understood. Initial research primarily focused on resin constituents like sesquiterpenes and triterpenes [5-9]. In 1951, King and colleagues discovered oligomeric resveratrols (oligomers of 3,5,4′-trihydroxystilbene) and hopeaphenol in *Hopea odorata* and *Balanocarpus heimii* [10, 11], shifting the research focus. Dipterocarp species produce various secondary metabolites, including volatile oils, triterpenoids, coumarins, flavonoids, quinones, phenolics, and resveratrol oligomers (oligostilbenoids) [12]. Notably, oligostilbenoids exhibit antibacterial, antiviral, and cytotoxic effects [13] and are unique to a limited number of plant families, including *Dipterocarpaceae*, *Cyperaceae*, *Gnetaceae*, *Leguminosae*, and *Vitaceae* [14].

Phytochemistry of Dipterocarpaceae

Non-oligomeric Compounds

Phytochemical investigations of dipterocarp plants initially analyzed resin constituents, revealing sesquiterpenes and triterpenes. In 1966, Diaz et al. [6] identified resin constituents in five species of the genus *Doona* (now *Shorea*), including *D. congestifolia*, *D. gardneri*, *D. macrophylla*, *D. oblonga*, and *D. zeylanica*, reporting four sesquiterpene derivatives: β-elemene (**1**), humulene (**2**), caryophyllene (**3**), and copaene (**4**). They also identified five triterpene derivatives: oleanene-type β-amyrin (**12**), dammarane-type dammarenediol-I (**13**), hydroxydammarenone-I (**14**), taraxastane-type Ψ-taraxasterol (**15**), and ursane-type ursolic acid (**16**). Further research by Bisset et al. [5] on 42 *Dipterocarpus* species revealed additional sesquiterpenes, including α-gurjunene (**5**), γ-gurjunene (**6**), alloaromadendrene (**7**), cyperene (**8**), calarene (**9**), farnesane (**10**), and dehydrofarnesane (**11**), along with triterpenes like dammaradienone (**17**), dipterocarpol (**18**), 20R/S-ocotillone (**19**), and dammarenediol-II (**20**). Additional investigations into *Anisoptera*, *Cotylelobium*, *Dryobalanops*, *Stemonoporus*, *Upuna*, and *Hopea* yielded compounds like those found in *Doona* and

Dipterocarpus [7, 15-17]. Other triterpenes identified in *Shorea* resin include ursolic aldehyde (**21**) [18], shoreic acid (**22**), and dammarenolic acid (**23**) [19]. In contrast, *Dryobalanops* resin yielded dryobalanonoloic acid (**24**), hedragonic acid (**25**), oleanolic acid acetate (**26**), dryobalanolide (**27**), and methyl-11-oxoasiatate (**28**) [17] (Figure 1).

The non-oligomeric constituents from other parts of Dipterocarpaceae are more varied than their resin constituents, including coumarins, flavonoids, and phenolic compounds. However, terpenoids remain common constituents of this family. In several studies, triterpenes found in the resin were also detected in the bark or timber extracts of dipterocarp plants, such as β-amyrin (**12**) [20], ursolic acid (**16**) [21, 20], dipterocarpol (**18**), and dammarenediol-II (**20**) [22,20]. On the other hand, several triterpene derivatives not identified in the resin were found in the bark or timber extracts, including lupane-type lupeol (**29**), betulinic acid (**30**), steroid-type sitosterol (**31**), and other derivatives such as oleanen-type δ-amyrenone (**32**) and ursane-type α-amyrin (**33**), as well as asiatic acid (**34**) [22,21,20,23]. This indicates that the terpene constituents of the non-resin parts are more varied, as they come not only from the dammaran, ursane, and oleanane groups but also from the lupane and steroid groups.

A study by Bate-Smith and Whitmore [3] reported the phenolic constituents of the leaves from 28 species across eight genera in the subfamily Dipterocarpoideae, including *Shorea*, *Hopea*, *Balanocarpus*, *Dipterocarpus*, *Dryobalanops*, *Anisoptera*, *Vatica*, and *Upuna,* through hydroxylation of the extracts followed by thin layer chromatography (TLC) observations. From this examination, five flavonoid derivatives were identified: myricetin (**35**), quercetin (**36**), kaempferol (**37**), delphinidin (**38**), and cyanidin (**39**); five cinnamic acid derivatives: ferulic acid (**40**), caffeic acid (**41**), sinapic acid (**42**), and p-coumaric acid (**43**); and a simple phenolic compound, especially an ellagic acid (**44**) constituent. Other investigations of the bark or timber from the genera *Stemonoporus, Shorea, Vatica, Vateria*, and *Cotylelobium*, using similar methods as reported by Bate-Smith and Whitmore, have also identified several phenolic compounds, such as benzoic acid derivatives like 4-hydroxybenzaldehyde (**45**) and methyl-2,4-dihydroxybenzoate (**46**); coumarin derivatives like scopoletin (**47**) and bergenin (**48**); the quinone derivative chrysophanol (**49**); and some ellagic acid derivatives [21,24,20].

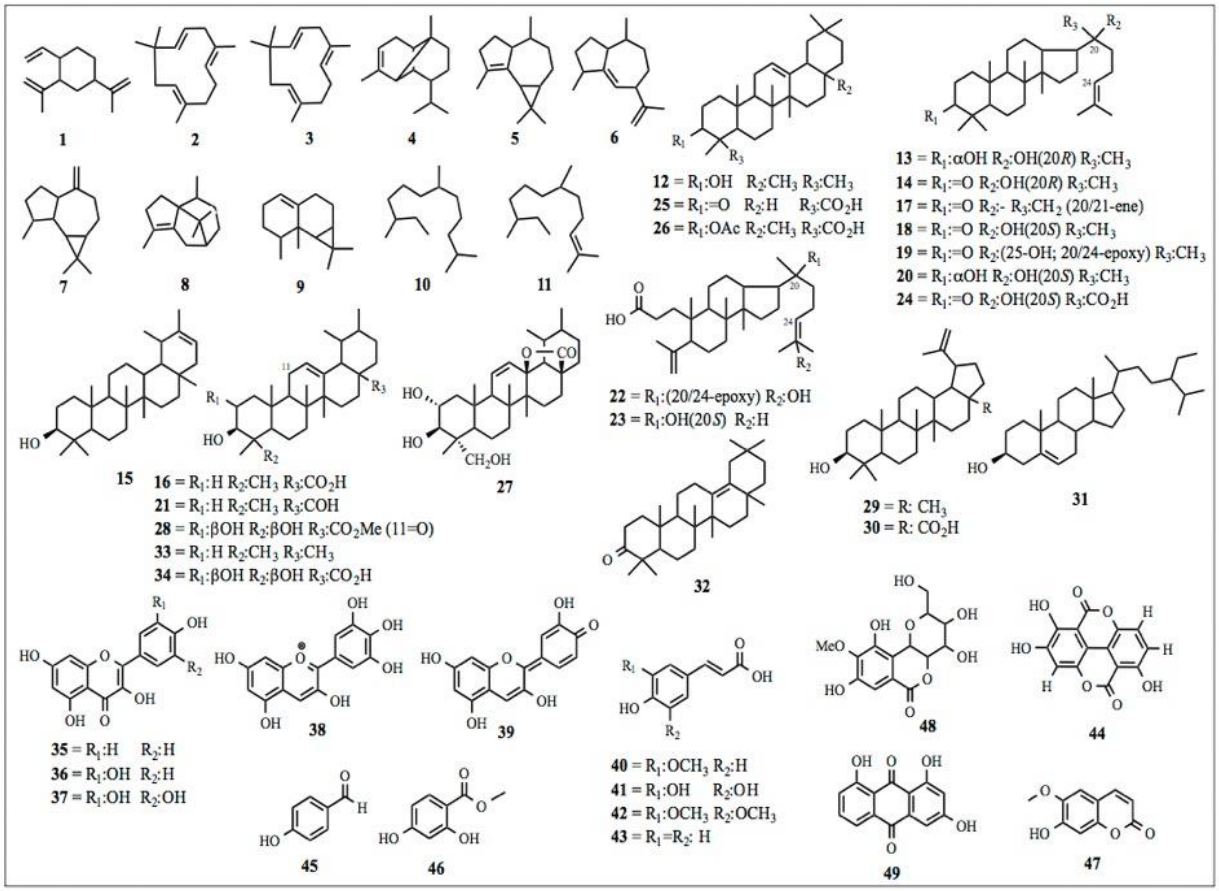

Figure 1. Structures of non-oligomeric compounds from the Dipterocarpaceae family.

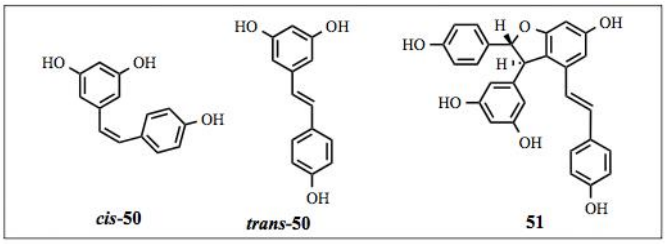

Figure 2. Structures of resveratrol (**50**) and ε-viniferin (**51**).

Oligomeric Compounds

Oligostilbenoids represent a unique class of biologically active natural products primarily produced by dipterocarp plants. The skeleton of oligostilbenoids is fundamentally based on a 14-carbon framework made up of two aromatic rings (phenol and resorcinol groups), which are linked by an ethylene bridge to create the resveratrol skeleton (**50**) (3,5,4′-trihydroxystilbene). The structure of **50** can form dimers, trimers, tetramers, or larger polymers, while their hydroxyl groups can be substituted with glucose, methyl, isoprene, or other groups. The chemical structures of oligostilbenoids are highly diverse, and to date, more than 300 oligostilbenoids have been isolated, with their structures clarified by modern spectroscopic methods [14]. The increasing numbers of oligostilbenoids are attributed to the degree of polymerization and the variety of polymerization patterns, stereochemistry, and derivatives.

Sotheeswaran and Pasupathy [12] previously reviewed and categorized oligostilbenoids into two major groups: group A, which includes oligostilbenoids containing at least one five-membered oxygen heterocyclic ring (1,2-dihydrobenzofuran ring), and group B, which consists of oligostilbenoids that do not contain any oxygen in their heterocyclic ring. In this classification, Sotheeswaran and Pasupathy propose that all naturally occurring compounds in group A are formed from resveratrol (**50**) through the dimer ε-viniferin (**51**) as the main precursor in the biogenetic pathway of oligostilbenoids (Figure 2). However, this biogenetic pathway is not always followed, as other biogenetic precursors may exist simultaneously during the biosynthetic process. This scheme has limited applicability, therefore we propose to reclassify oligostilbenoids in the Dipterocarpaceae based on the characteristics of their carbon skeletons in each group, such as the presence of benzofuran and bicyclo ring systems in their structures.

Resveratrols

Resveratrol (**50**) was discovered in 1940 from the roots of the hellebore lily, *Veratrum grandiflorum* (Melathiaceae), by Takaoka [25]. In 1963, it was re-isolated from the roots of *Polygonum cuspidatum* (Polygonaceae), a traditional Chinese and Japanese medicinal plant “Ko-jo-kon” [26]. However, **50** did not attract any interest until 1992, when it was discovered to be a component of red wine and was implicated in the "French paradox," an epidemiological finding that highlights an inverse relationship between heart disease incidence and red wine consumption [27]. Recently, **50** has been identified in several families, including *Vitaceae* and *Moraceae*; however, it has only been reported from four species of Dipterocarpaceae, namely *Vatica rassak* [28,29], *Hopea utilis* [30], *H. exalata* [31], and *Vateria indica* [32]. As an active compound and precursor to all oligostilbenoids, **50** is found in only a few species and exists in plants as its derivatives (Table 1).

Table 1. Distribution of resveratrol and its derivatives in the Dipterocarpaceae family.

No	Compounds	Sources
50	Resveratrol	*Vatica rassak* [28], *Hopea utilis* [30], *H. exalata* [31], and *Vateria indica* [32]
52	*trans*-Piceid	*Vatica diospyroides* [33], *V. rassak* [28], *V. albiramis* [34], *Vateria indica* [35], *Upuna borneensis* [36], *Cotylelobium lanceolatum* [37], *Hopea hainanensis* [38], and *H. parviflora* [39]
53	*cis*-Piceid	*Vatica diospyroides* [40] and *Upuna borneensis* [41]
54	Piceid-2"-*O*-coumarate	*U. borneensis* [36]
55	Piceid-2'-*O*-*p*-hydroxybenzoate	*U. borneensis* [36]
56	Piceid-2'-*O*-*E*-ferulate	*U. borneensis* [36]
57	Resveratrol-3-*C*-β-glucopyranoside	*Shorea hemsleyana* [42] and *S. seminis* [43]
58	Resveratrol-2-*C*-β-glucopyranoside	*Hopea utilis* [30] and *Vateria indica* [32]

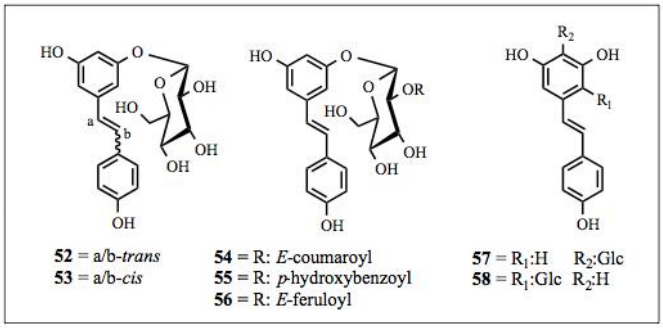

Figure 3. Structures of resveratrol derivatives from the Dipterocarpaceae family.

Piceid (**52**) is the first resveratrol glucoside derivative reported from plants. This compound was isolated from the leaves of *Picea glehnii* (Pinaceae) by Kariyone et al. [44]. Its structure was established a year later as *trans*-3,5,4′-trihydroxystilbene-3-*O*-β-D-glucopyranoside based on infrared and ultraviolet spectral data of oxidative decomposition of its methylated derivative [45]. This compound was reported in several species of Dipterocarpaceae, such as *Vatica diospyroides* [33], *V. rassak* [28], *V. albiramis* [34], *Vateria indica* [35,32], *Upuna borneensis* [36,41], *Cotylelobium lanceolatum* [37], *Hopea hainanensis* [38], *H. utilis* [30], and *H. parviflora* [39]. Its isomer, *cis*-piceid (**53**), was reported from *Vatica diospyroides* [40] and *Upuna borneensis* [41]. Other derivatives of **52**, such as *O*-glucoside types (glucosylated at hydroxyl group), piceid-2''-*O*-coumarate (**54**), piceid-2'-*O*-*p*-hydroxybenzoate (**55**), and piceid-2'-*O*-*E*-ferulate (**56**), have been reported from *U. borneensis* [36]. In contrast, its *C*-glucoside types (glucosylated at aromatic carbon), resveratrol-3-*C*-β-glucopyranoside (**57**) was reported from *Shorea hemsleyana* [42] and *S. seminis* [43], and resveratrol-2-*C*-β-glucopyranoside (**58**) from *Hopea utilis* [30] and *Vateria indica* [32] (Figure 3).

Dimer Oligostilbenoids

Several genera within the Dipterocarpaceae family, including six from the Dipterocarpeae tribe (*Stemonoporus, Cotylelobium, Dipterocarpus, Upuna, Vateria,* and *Vatica*) and four from the Shoreae tribe (*Hopea, Balanocarpus, Neobalanocarpus,* and *Shorea*), have been reported to have a variety of dimer oligostilbenoids (Table 2). The most prevalent of these is (-)-ε-viniferin (**51**), initially identified in the leaves of *Vitis vinifera* (Vitaceae) [46]. Characterized as phytoalexins in oligomeric forms of trihydroxy stilbenes, this dimer is proposed as a precursor for all oligostilbenoids containing a benzofuran ring [12]. In the Dipterocarpaceae family, **51** was discovered in *Stemonoporus affinis* [47]. The dimer oligostilbenoids from dipterocarp plants exhibit diverse carbon skeletons, which can be categorized into five main groups:

A. Dimer oligostilbenoids with a dihydrobenzofuran ring and free stilbene skeleton.

B. Dimer oligostilbenoids with a dihydrobenzofuran ring and dibenzocycloheptane ring.

C. Dimer oligostilbenoids with a dihydrobenzofuran ring and dibenzocyclohexane ring.

D. Dimer oligostilbenoids from group B degradation, losing a p-hydroxybenzyl group.

E. Dimer oligostilbenoids without dihydrobenzofuran rings.

Trans-(-)-ε-viniferin (**51**) is the common dimer oligostilbenoid from group A, reported in several Dipterocarpaceae genera, including *Stemonoporus, Vatica, Shorea, Dipterocarpus, Hopea, Upuna, and Cotylelobium* [47,48,28,30,49,50]. Its stereoisomers include *cis*-(-)-ε-viniferin (**60**) from *Vateria indica* and *V. albiramis* [34,32]. Both trans- (59) and *cis*-(+)-ε-viniferins (61) were identified from *Cotylelobium lanceolatum* [51,52]. This dimer serves as a key precursor in the biogenetic pathways of other oligostilbenoids, particularly those with a benzofuran ring [12]. Other oligostilbenoids in this group include glucoside derivatives, like resveratrol (**50**). Two types of glucoside derivatives belonging to *O*-glucosides and *C*-glucosides have been identified. For *O*-glucosides, paucifloroside A (**62**) from *Vatica pauciflora* [53] and its stereoisomer upunoside D (**63**) from *Upuna borneensis* [54] have been identified. As for *C*-glucosides, (-)-diptoindonesin A (**64**) from *Shorea seminis* [43] and uliginoside A (**65**) and hemsleyanoside B (**66**) from *S. uliginosa* [55] have also been reported. Additionally, (-)-laevifonol (**67**) from *S. laeviforia* [56] and its stereoisomer shorealactone (**68**) from *S. hemsleyana* [49] also belong to this group despite their vinyl groups being substituted by ascorbic acid (Figure 4).

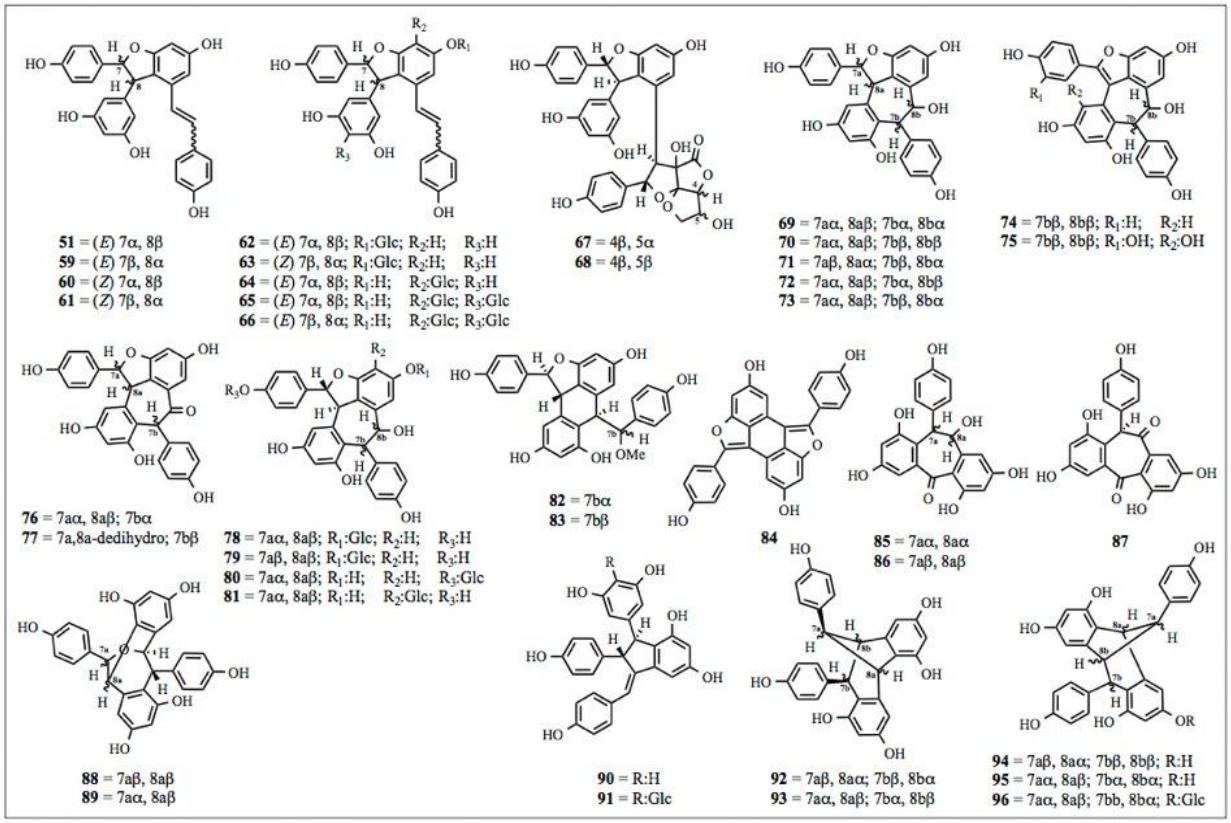

Figure 4. Structures of dimer oligostilbenoids from the Dipterocarpaceae family.

In group B, (-)-balanocarpol (**69**), featuring a dibenzocycloheptane ring as its main skeleton, has been reported from *Balanocarpus zeylanicus* and *Hopea jucunda* [57]. Its stereoisomer, (+)-balanocarpol (**70**), was identified in *H. parviflora* and *H. hainanensis* [58,38]. With four chiral carbons, **69** can form 16 stereoisomers. Yet, only three others have been documented from Dipterocarpaceae: (-)-ampelopsin A (**71**) from *H. parviflora* [58], (+)-ampelopsin A (**72**) from *H. utilis* [30], and hemsleyanol A (**73**) from *Shorea hemsleyana* [42]. Several derivatives have also been reported, including hydroxylated, oxidized, and glucosylated forms. Hydroxylated derivatives Malibatols A (**74**) and B (**75**) were derived from the leaves of *Hopea malibatol* [59]. Oxidized derivatives, such as pauciflorol E (**76**) and shoreaphenol (**77**), come from the stem barks of *Vatica pauciflora* and *Shorea robusta* [60], respectively. Glucoside derivatives include laevifoside (**78**) from *Shorea laeviforia* [56], vatalbinosides D (**79**) and E (**80**) from *Vatica albiramis* [34], and hemsleyanoside (**81**) from *Shorea hemsleyana* [61].

In group C, oligostilbenoids that contain a dibenzocyclohexane ring in their skeleton were reported only from two species: *Welwitschia mirabilis* (*Welwitschiaceae*) [62] and *Hopea hainanensis* (Dipterocarpaceae) [38]. Hopeahainols E (**82**) and F (**83**) and their dedihydro derivative, hopeahainol C (**84**), from the stem wood of *H. hainanensis* [38], are oligostilbenoids from this group that have been reported from plants. In group D, there are only three compounds, that are formed from the degradation reaction of group B: parviflorol (**85**) from *Hopea parviflora*, *H. dryobalanoides*, and *Shorea hopeifolia* [58,63,64]; hemsleyanol E (**86**) from *S. hemsleyana* [61]; and diptoindonesin D (**87**) from *Hopea dryobalanoides* [63].

In group E, (+)-ampelopsin D (**90**) from *Vatica pauciflora* [53] and its glucoside derivative, hemsleyanoside A (**91**), from *Shorea hemsleyana* [65] are dimer oligostilbenoids containing a unit of a benzopentacyclic ring in their structures. Meanwhile, (-)-ampelopsin F (**92**) and (-)-isoampelopsin F (**94**) from *Vatica pauciflora* [53], (+)-ampelopsin F (**93**) and (+)-isoampelopsin F (**95**) from *Cotylelobium melanoxylon* [52], and its C-glucoside derivative, upunoside C (**96**), from *Upuna borneensis* [54] contain a dibenzobicyclo[3.2.1]octadiene ring. Additionally, (-)-heimiol A (**88**) and hopeahainol D (**89**) from the genera *Neobalanocarpus* and *Hopea* [66,67,38] can also be classified in this group. Both compounds possess a bicycloheterocyclic ring: 6-oxadibenzobicyclo[3.2.2]nonadiene.

Table 2. Distribution of dimer oligostilbenoids in the Dipterocarpaceae family.

No	Compounds	Sources
51	*trans*-(-)-ε-Viniferin	*Stemonoporus affinis* [47], *Vatica rassak* [28], *V. pauciflora* [53], *V. umbonata* [68], *V. albiramis* [34], *Shorea disticha* [48], *S. laeviforia* [56], *S. hopeifolia* [64], *Dipterocarpus grandiflorus* [49], *D. hasseltii* [69], *Hopea utilis* [30], *H. parviflora* [58], *Upuna borneensis* [34], *Cotylelobium lanceolatum* [50], *Dryobalanops aromatica* [70], *D. lanceolata* [71], *D. rappa* [72], and *D. beccarii* [73]
59	*trans*-(+)-ε-Viniferin	*C. melanoxylon* [51]
60	*cis*-(-)-ε-Viniferin	*Vateria indica* [32] and *V. albiramis* [34]
61	*cis*-(+)-ε-Viniferin	*Cotylelobium melanoxylon* [52]
62	Paucifloroside A	*Vatica pauciflora* [53], *Upuna borneensis* [54], and *Vateria indica* [32]
63	Upunoside D	*Upuna borneensis* [54] and *Vateria indica* [32]
64	(-)-Diptoindonesin A	*Shorea seminis* [43], *Dryobalanops aromatica*[70], and *D. beccarii* [73]
65	Uliginoside A	*S. uliginosa* [55]
66	Hemsleyanoloside B	*S. hemsleyana* [65] and *S. uliginosa* [55]
67	(-)-Laevifonol	*S. laeviforia* [56], *S. balangeran* [74], *Vatica umbonata* [68] and *Dipterocarpus hasseltii* [69], *Dryobalanops aromatica* [70], *D. lanceolata* [71], and *D. rappa* [72]
68	Shorealactone	*Shorea hemsleyana* [75] and *Dipterocarpus grandiflorus* [49]
69	(-)-Balanocarpol	*Hopea jucunda* and *Balanocarpus zeylanicus* [57], *H. malibato* [59], *H. parviflora* [58], *H. utilis* [76], *H. dryobalanoides* [63], *Shorea roxburghii* [77], *Neobalanocarpus heimii* [66], *Vateria indica* [30], and *Vatica albiramis* [34]
70	(+)-Balanocarpol	*Hopea parviflora* [58] and *H. hainanensis* [38]
71	(-)-Ampelopsin A	*H. parviflora* [58], *Shorea seminis* [43], *S. laeviforia* [56], *S. gibbosa* [78], *Dipterocarpus grandiflorus* [49], and *Vatica albiramis* [34]
72	(+)-Ampelopsin A	*Hopea utilis* [30] and *Vateria indica* [76]
73	Hemsleyanol A	*Shorea hemsleyana* [42], *Vatica pauciflora* [53], and *S. balangeran* [74]
74	Malibatol A	*Hopea malibato* [59], *H. utilis* [30], *H. hainanensis* [38], *H. parviflora* [39], and *Vateria indica* [32]
75	Malibatol B	*Hopea malibato* [59]
76	Pauciflorol E	*Vatica pauciflora* [60]
77	Shoreaphenol/ Hopeafuran	*Shorea robusta* [79], *Hopea utilis* [30], *H. dryobalanoides* [63], and *H. exalata* [31]
78	Laevifoside	*Shorea laeviforia* [56]
79	Vatalbinoside D	*Vatica albiramis* [34]
80	Vatalbinosides E	*V. albiramis* [34]
81	Hemsleyanoside F	*Shorea hemsleyana* [61], *S. laeviforia* [56], and *S. parvistipulata* [80]
82	Hopeahainol E	*Hopea hainanensis* [38]
83	Hopeahainol F	*H. hainanensis* [38]
84	Hopeahainol C	*H. hainanensis* [38]
85	Parviflorol	*H. parviflora* [58], *H. dryobalanoides* [63], and *Shorea hopeifolia* [64]
86	Hemsleyanol E	*S. hemsleyana* [61]
87	Diptoindonesin D	*Hopea dryobalanoides* [63]
88	(-)-Heimiol A	*Neobalanocarpus heimii*[66], *Hopea dryobalanoides*[63], *H. mengarawan* [68], and *H. hainanensis* [38]
89	Hopeahainol D	*H. hainanensis* [38]
90	(+)-Ampelopsin D	*Vatica pauciflora* [53]
91	Hemsleyanoside A	*Shorea hemsleyana* [65]
92	(-)-Ampelopsin F	*Vatica pauciflora* [53], *V. umbonata* [81], *Vateria indica* [32], *Dipteroearpus grandiflorus* [49], and *Upuna* borneensis [82]
93	(+)-Ampelopsin F	*Cotylelobium melanoxylon* [52], *Dryobalanops lanceolata* [71], and *D. rappa* [72]

94	(-)-Isoampelopsin F	*Vatica pauciflora* [53], *Dipteroearpus grandiflorus* [49], and *Upuna borneensis* [82]
95	(+)-Isoampelopsin F	*Cotylelobium melanoxylon* [52], and *Dryobalanops rappa* [72]
96	Upunoside C	*Upuna borneensis* [54]

Trimer oligostilbenoids

The trimer oligostilbenoid structure includes an additional resveratrol unit (**50**) compared to the dimer oligostilbenoid, resulting in greater variability in its skeleton types. According to Sotheeswaran and Pasupathy [12], oligostilbenoids with a benzofuran ring are biogenetically derived from ε-viniferin (**51**), suggesting that these trimer oligostilbenoids form from the condensation of units **50** and **51**. They can be classified into four main groups based on the number of benzofuran rings:

A. Trimer oligostilbenoids with three benzofuran rings.

B. Trimer oligostilbenoids with two benzofuran rings.

C. Trimer oligostilbenoids with one benzofuran ring.

D. Trimer oligostilbenoids without benzofuran rings.

In group A, two compounds were highlighted: (+)-α-viniferin (**97**), sourced from *Hopea exalata* [31], and its stereoisomer, (-)-α-viniferin (**98**), derived from *Shorea seminis* [43] (Figure 5). Notably, the oligostilbenoids within this group are quite limited and lack diversity despite the presence of six chiral carbons and six hydroxyl groups in their structures. This suggests that the skeleton type of this group originates from restricted precursors, and the conformation of its skeleton restricts the entry of larger groups, such as glucose molecules. Currently, only four derivatives of compound **97** have been reported: the *O*-glucoside derivative, (+)-α-viniferin-13β-*O*-D-glucopyranoside (**99**) from *S. hemsleyana* [42]; oxidative derivatives, hopeanolin (**100**) from *Hopea exalata* [31] and grandiphenol C (**101**) from *Dipterocarpus grandiflorus*; and a rearranged derivative, grandiphenol D (**102**) also from *D. grandiflorus* [83].

Group B contains only a few reported oligostilbenoids with two units of dihydrobenzofuran rings. Notable examples include trans-ampelopsin E (**103**) from *Shorea gibbosa* [78] and *S. hopeifolia* [64], along with miyabenol C (**104**) from *Dipterocarpus grandiflorus* [49], both retaining a free stilbene skeleton. In contrast, Hemsleyanol B (**105**) from *Shorea hemsleyana* [42] and *S. balangeran* [74] features cyclized stilbene skeletons. Other oligostilbenoids exhibit uncommon benzofuran rings: the second benzofuran rings in trans- (**106**) and cis-diptoindonesins B (**107**) from *Dryobalanops oblongifolia* [84], as well as uliginosides B (**109**) and C (**110**) from *Shorea uliginosa* [55], arising from C3-C8 type condensation. Melanoxylin B (**108**) from *Cotylelobium melanoxylon* exhibits C8-C12 type condensation. Such condensations are also common in families beyond *Dipterocarpaceae,* including *Vitaceae, Gnetaceae,* and *Fabaceae* [14].

The number of oligostilbenoids qualifying for group C is larger and more diverse than in the other groups. This suggests that group C represents the primary set of compounds within Dipterocarpaceae. Based on the types of ring systems in their structures, the oligostilbenoids in group C can be categorized into four subgroups, which are:

C1. Trimer oligostilbenoids with a dibenzobicyclo [5.3.0]decadiene ring system,

C2. Trimer oligostilbenoids with a tribenzobicyclo [6.3.0]undecadiene ring system,

C3. Trimer oligostilbenoids with a dibenzobicyclo [3.3.0]octadiene ring system, and

C4. Trimer oligostilbenoids with a benzocyclopentane ring.

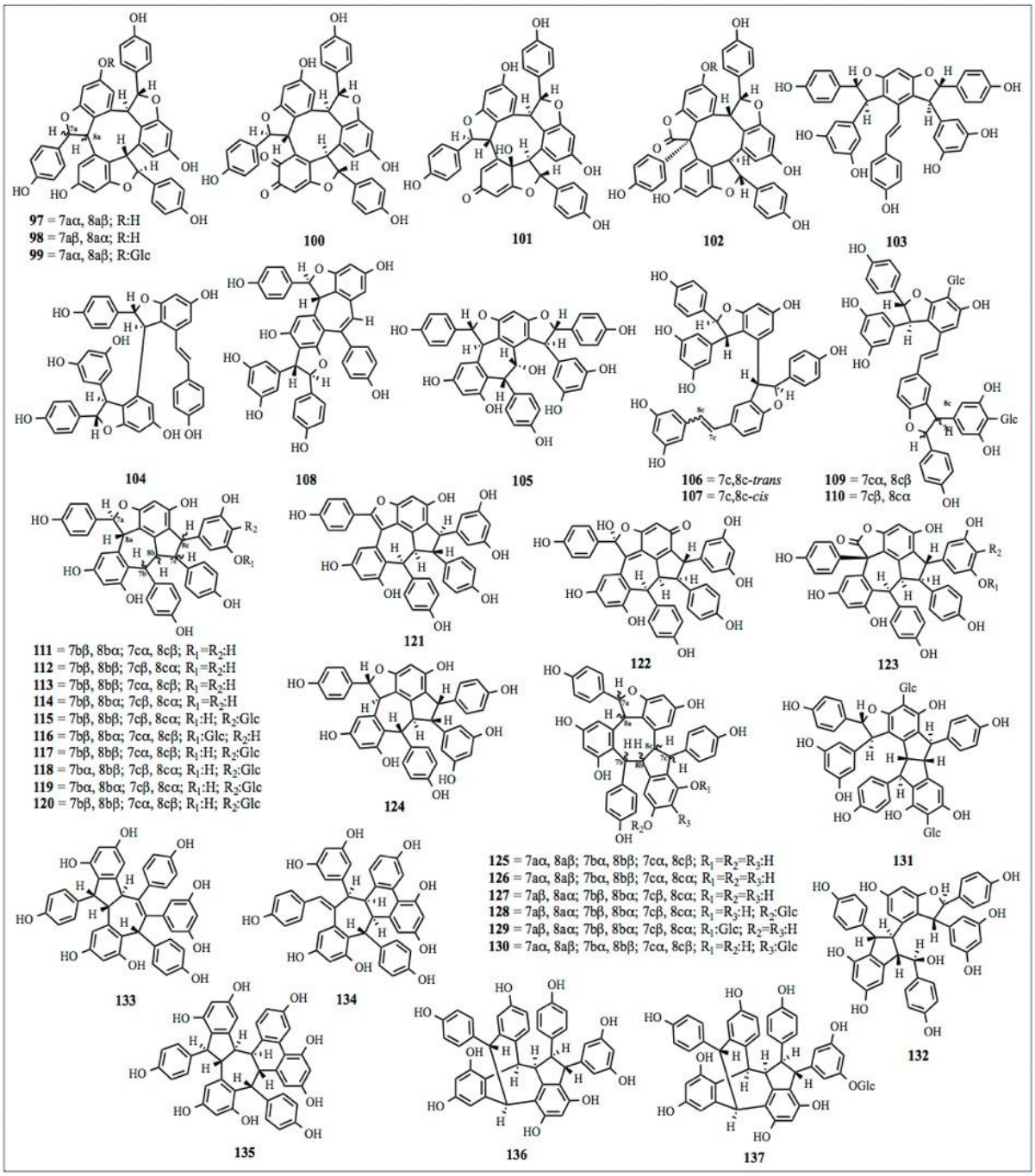

Figure 5. Structures of trimer oligostilbenoids from the Dipterocarpaceae family.

In subgroup C1, 14 oligostilbenoids have been identified that contain a bicyclo ring system: dibenzobicyclo[5.3.0]decadiene, featuring vaticanol A (**111**) from *Hopea exalata* [31], pauciflorols A (**112**) and B (**113**) from *Vatica pauciflora* [53], along with vaticanol E (**114**) from *V. rassak* [85]. In addition to these stereoisomers, their glucoside derivatives have also been documented, such as the *O*-glucoside derivative vaticaside A (**116**) from *V. rassak* [85] and *C*-glucoside derivatives, including hemsleyanoside E (**115**) from *Shorea hemsleyana* [42], melapinol A (**117**) from *Shorea sp*. [86], upunoside B (**118**) from *Upuna borneensis*[54], as well as cotylelosides A (**119**) and B (**120**) from *Cotylelobium lanceolatum* [37]. Melapinol B (**121**) from *Hopea exalata* [31] and cotylelophenol B (**122**) from *Cotylelobium lanceolatum* [50] are 7a,8a-dedihydro and oxidative derivatives of **113**, respectively, while cotylelophenol A (**123**) from *C. lanceolatum* [50] is the rearranged product of **112** (Figure 5). Moreover, davidiol A (**124**), isolated from *Shorea hemsleyana* [42], also qualifies for this group, but the positions of rings C1 and C2 are opposite compared to the other derivatives. Besides that, in subgroup C2, three oligostilbenoids containing the tribenzobicyclo[6.3.0]undecadiene ring system are identified: distichol (**125**) from *Shorea disticha* [48]

and *Sternonoporus canaliculatus* [87], canaliculatol (**126**) from *S. canaliculatus* [87], and stenophyllol B (**127**) from *Vatica pauciflora* [53] and *V. umbonata* [81]. Their glucoside derivatives include pauciflorosides B (**128**) and C (**129**) from *V. pauciflora* [53], along with hemsleyanoside D (**130**) from *Shorea hemsleyana* [65]. Subgroup C3 features a single oligostilbenoid, hemsleyanoside C (**131**), isolated from the bark of *S. hemsleyana* [65], which contains a dibenzobicyclo [3.3.0]octadiene ring. Lastly, subgroup C4 contains only davidiol B (**132**), obtained from the stem bark of *Vatica pauciflora* [53], which includes a cyclopentane ring.

In group D, copalliferol B (**133**) from *Vateria copallifera* [88] features a bicyclo ring system, specifically dibenzobicyclo[5.3.0]decatriene. In contrast, stemonoporol (**134**), isolated from various *Stemonoporus* species, contains a tribenzobicyclo[5.4.0]undecatriene ring [47]. Additionally, copalliferol A (**135**) from *Vateria copallifera* possesses a tricyclo ring system, tetrabezotricyclo[8.4.0.$0^{2,6}$]tetradecatetraene [89]. Meanwhile, vaticanol G (**136**) from *Hopea exalata* [31] and vaticaside D (**137**) from *Vatica rassak* [85] both exhibit a tribezotricyclo[6.3.2.$0^{2.6}$] tetradecatetraene ring.

The distribution of all trimer oligostilbenoids in Dipterocarpaceae is summarized in Table 3. *Shorea* species are identified as the primary source, with 18 reported compounds, followed by *Vatica* (11 compounds), *Cotylelobium* (11 compounds), and *Hopea* (8 compounds). Furthermore, the types of oligostilbenoids isolated from *Shorea* are more diverse and representative of all groups (A-D), while those from *Vatica*, *Cotylelobium*, and *Hopea* are more specific to group C.

Table 3. Distribution of trimer oligostilbenoids in the Dipterocarpaceae family.

No.	Compounds	Sources
97	(+)-α-viniferin	*Dipterocarpus grandiflorus* [49], *Hopea dryobalanoides* [63], *H. exalata* [31], *Shorea hemsleyana* [42], *S. roxburghii* [77], and *Dryobalanops aromatica* [70]
98	(-)-α-viniferin	*Shorea seminis* [43], *S. gibbosa* [78], and *Dipterocarpus hasseltii* [69]
99	(+)-α-viniferin-13β-*O*-β-glucopyranoside	*Shorea hemsleyana* [42]
100	Hopeanolin	*Hopea exalata* [31]
101	Grandiphenol C	*Dipterocarpus grandiflorus* [83]
102	Grandiphenol D	*D. grandiflorus* [83]
103	*trans*-Ampelopsin E	*Shorea gibbosa* [78], *S. hopeifolia* [64], *Dryobalanops aromatica* [70], *D. rappa* [72], and *D. beccarii* [73]
104	Miyabenol C	*Dipterocarpus grandiflorus* [49]
105	Hemsleyanol B	*Shorea hemsleyana* [42] and *S. balangeran* [74]
106	*trans*-diptoindonesin B	*Dryobalanops oblongifolia* [84]
107	*cis*-diptoindonesin B	*D. oblongifolia* [84]
108	Melanoxylin B	*Cotylelobium melanoxylon* [52]
109	Uliginoside B	*Shorea uliginosa* [55]
110	Uliginoside C	*S. uliginosa* [55]
111	Vaticanol A	*Hopea exalata* [31], *Hopea hainanensis* [38], *Shorea sp.* [86], *S. bablangeran* [90], *Cotylelobium lanceolatum* [50], *C. melanoxylon* [52], and *Vatica rassak* [28]
112	Pauciflorol A	*Vatica pauciflora* [53], *Cotylelobium lanceolatum* [50], *Hopea exalata* [31], and *Vateria indica* [32]
113	Pauciflorol B	*Vatica pauciflora* [53]
114	Vaticanol E	*V. rassak* [85], *Cotylelobium melanoxylon* [52], and *Hopea hainanensis* [38]
115	Hemsleyanoside E	*Shorea hemsleyana* [65]
116	Vaticaside A	*Vatica rassak* [85] and *Cotylelobium lanceolatum* [37]
117	Melapinol A	*Shorea sp.* [86]
118	Upunoside B	*Upuna borneensis* [54]
119	Cotyleloside A	*Cotylelobium lanceolatum* [37]
120	Cotyleloside B	*C. lanceolatum* [37]
121	Melapinol B	*Hopea exalata* [31] *and Shorea sp.* [86]

122	Cotylelophenol B	*Cotylelobium lanceolatum* [50]
123	Cotylelophenol A	*C. lanceolatum* [50]
124	Davidiol A	*Shorea hemsleyana* [42]
125	Distichol	*S. disticha* [48] *and Sternonoporus canaliculatus* [87]
126	Canaliculatol	*S. canaliculatus [87]*
127	Stenophyllol B	*Vatica pauciflora* [53] *and V. umbonata* [81]
128	Paucifloroside B	*V. pauciflora* [53]
129	Paucifloroside C	*V. pauciflora* [53]
130	Hemsleyanoside D	*Shorea hemsleyana* [65]
131	Hemsleyanoside C	*S. hemsleyana* [65]
132	Davidiol B	*Vatica pauciflora* [53]
133	Copalliferol B	*Vateria copallifera* [88]
134	Stemonoporol	*Stemonoporus affinis, S. cordifolius, S. elegans, S. kanneliensis, S. lancifolius, S. oblongifolius* and *S. canaliculatus* [47]*, Vateria copallifera* [87], and *Shorea stipularis* [91]
135	Copalliferol A	*Vateria copallifera* [89,91]*, Stemonoporus affinis, S. cordifolius, S. elegans, S. kanneliensis, S. lancifolius, S. oblongifolius* and *S. canaliculatus* [47]*, Hopea cordifolia* [87]*, H. brevipetiolaris* and *Shorea stipularis* [91], and *Neobalanocarpus heimii* [66]
136	Vaticanol G	*Hopea exalata* [31]*, H. Mengarawan* [68]*, Vatica rassak* [85]*, V. pauciflora* [53]*, V. umbonata* [81]*, Shorea balangeran* [90]*, Cotylelobium lanceolatum* [50], and *C. melanoxylon* [52]
137	Vaticaside D	*Vatica rassak* [85]*, Vatica pauciflora* [53], and *Cotylelobium lanceolatum* [37]

Tetramer oligostilbenoids

The ε-viniferin skeleton in tetramer oligostilbenoids is identified by the presence of benzofuran rings. Most tetramer oligostilbenoids from dipterocarp plants feature two benzofuran units, indicating their biogenetic origin from two ε-viniferin (**51**) molecules. Numerous tetramer oligostilbenoids are currently reported from various Dipterocarpaceae species, classified into six distinct groups based on their skeletal types.

A. Tetramer oligostilbenoids with two symmetrical dibenzocycloheptane rings,

B. Tetramer oligostilbenoids with a dibenzobicyclo [5.3.0]decadiene ring system,

C. Tetramer oligostilbenoids with a dibenzobicyclo [3.3.0]nonadiene ring system,

D. Tetramer oligostilbenoids with a dibenzobicyclo [3.2.1]nonadiene ring system,

E. Tetramer oligostilbenoids with tetrahydrofuran ring, and

F. Others.

The oligostilbenoids of group A consist of two dibenzocycloheptane rings formed through the condensation of two molecules of **51** [92,93]. The first oligostilbenoid discovered in plants was (-)-hopeaphenol (**138**), which was isolated from *Balanocarpus heimii* and *Hopea odorata* by Coggon et al. [10]. Its structure was determined a year later through X-ray crystallography of its bromide derivative [94,11]. The presence of eight chiral carbons in **138** allows for numerous stereoisomers, with nine reported from Dipterocarpaceae: (-)-(**139**) and (+)-isohopeaphenols (**140**) from *Shorea hemsleyana* [65,42], hopeaphenol A (**141**) and isohopeaphenol A (**142**) from *Vatica oblongifolia* [34], pauciflorol C (**143**) from *V. pauciflora* [53], vateriaphenols B (**144**) and D (**145**) from *V. indica* [35,95], as well as neohopeaphenol A (**146**) and isoneohopeaphenol A (**147**) from *Hopea hainanensis* [67]. Additionally, glucoside derivatives such as vaterioside B (**148**) from *Vateria indica* [32], vatalbinoside A (**149**) from *Hopea parviflora* [39], and diptoindonesin F (**150**) from *Shorea gibbosa* [78] have been noted, alongside oxidative derivatives like vateriaphenol D (**151**) from *Vateria indica* [76], dibalanocarpol (**152**) from *Hopea malibato* [59], stenophyllol A (**153**), and upunaphenol B (**155**) from *Upuna borneensis* [96], as well as pauciflorol B (**154**) from *Vatica pauciflora* [53]. Upunaphenol I (**156**) from *Upuna borneensis* [34] is also part of this group, though its structure is altered by adding a carbon bridge connecting two aromatic rings B2/C2 via carbons C14b and C14c. Three derivatives from *U. borneensis*, upunaphenols H (**157**) [34] and P (**158**) [97] are formed through the oxidation of **156** at their p-hydroxybenzene and aliphatic rings, respectively, and upunaphenol J (**159**) [34] is derived from the degradation of carbons C8b and C8c, which links the two cycloheptane ring units of **156** (Figure 6).

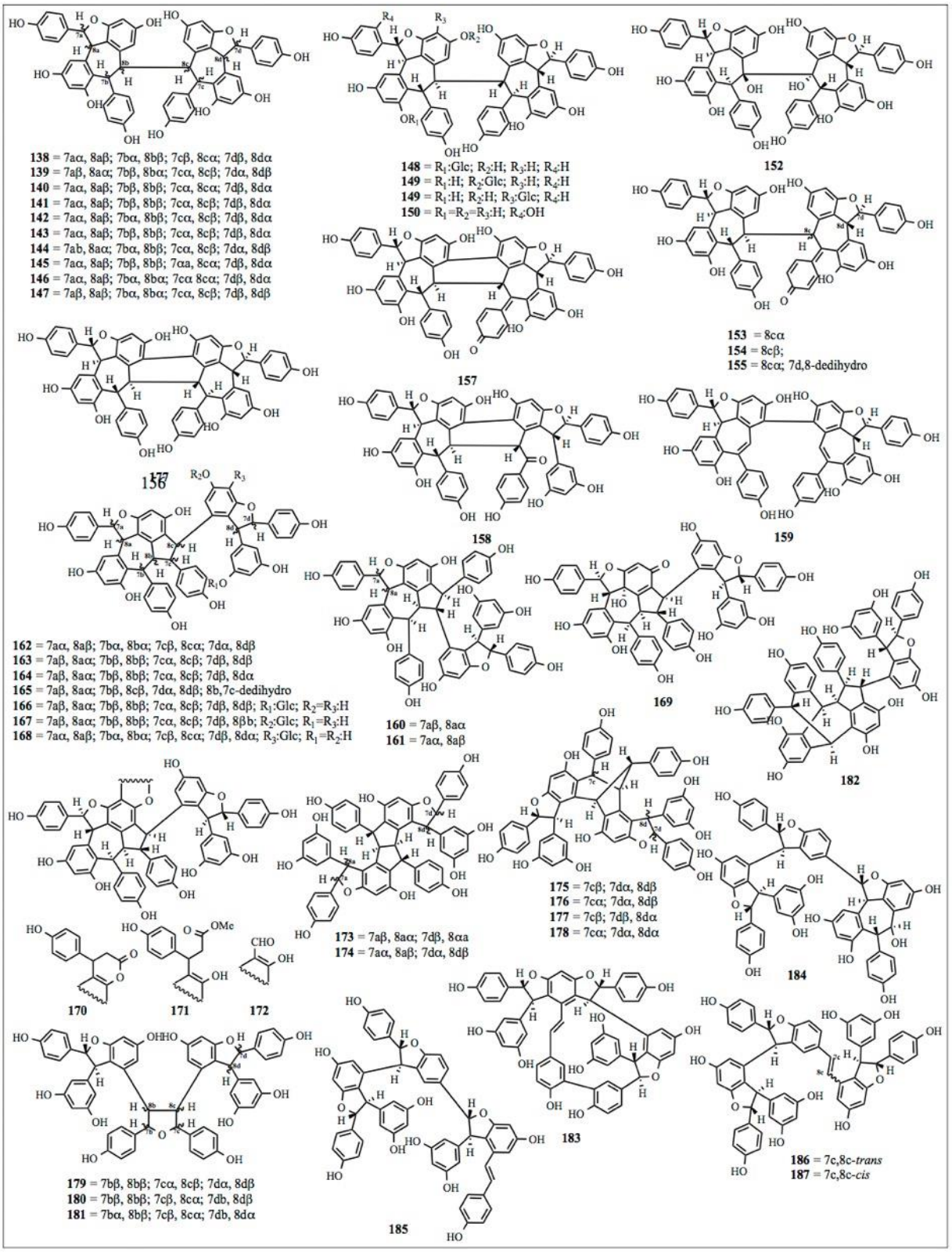

Figure 6. Structures of tetramer oligostilbenoids from the Dipterocarpaceae family.

In group B, two types of oligostilbenoids are identified based on their condensation schemes. The first type, the C8-C8 type, features a cyclopentane ring formed from carbons C8b and C8c in the dibenzobicyclo[5.3.0]decadiene system. The second type, the C8-C14 type, results from the condensation of carbons C8b and C14c. While the bicyclo ring systems in both types resemble those of trimer oligostilbenoids from subgroup C1, they differ in their precursor origins. The pentacyclic ring in the first type

exhibits high steric hindrance, leading to the report of only two compounds, hemsleyanols C (**160**) and D (**161**) from *Shorea hemsleyana* [61]. In contrast, the second type has three reported stereoisomers: vaticaphenol A (**162**) from *Vatica diospyroides* [33], vaticanol B (**163**) from *V. rassak* [28], and isovaticanol B (**164**) from *V. pauciflora* [53]. Additionally, oxidative derivatives such as upunaphenol G (**165**) and upunaphenol F (**169**) from *Upuna borneensis* [41], along with glucoside derivatives vaticasides B (**166**) and C (**167**) from *Vatica rassak* [85] and vatalbinoside B (**168**) from *V. albiramis* [34], have been reported. Other less common types include upunaphenols L (**170**), M (**171**), and N (**172**) from *Upuna borneensis* [98].

Very few oligostilbenoids are classified in groups C, D, and E, and glucoside or oxidative derivatives have not been reported from Dipterocarpaceae. In group C, only two oligostilbenoids exhibit a bicyclo ring system: dibenzobicyclo[3.3.0]nonadiene, namely (-)-ampelopsin H (**173**) from *Shorea hemsleyana* [61] and stenophyllol C (**174**) from *Upuna borneensis* [96]. Group D includes four stereoisomers: vatdiospyroidol (**175**) from *Vatica diospyroides* [33], vaticanols C (**176**) [28] and F (**178**) [85] from *V. rassak*, and isovaticanol C (**177**) from *V. pauciflora* [53]. The bicyclo ring system dibenzobicyclo[3.2.1]nonadiene in this group is structurally similar to the main skeleton of dimer (+)-ampelopsin F (**93**), suggesting analogous biogenetic pathways, differing only in the precursor used for oligomerization. In group E, a tetrahydrofuran ring acts as the main skeleton, formed from the condensation of **51** and its epoxide derivative. This group includes three stereoisomers: grandiphenols A (**179**) and B (**180**) from *Dipterocarpus grandiflorus* [49] and vateriaphenol F (**181**) from *Vateria indica* [32].

Group F includes oligostilbenoids that could not be classified in groups A-E, such as cotylelophenol C (**182**) from *Cotylelobium lanceolatum* [37], characterized by a tricyclo ring system of tetrabezotricyclo[8.4.0.$0^{2,6}$] tetradecatetraene as the main skeleton. This tricyclo ring system is similar to trimer vaticanol G (**136**). Besides that, some unusual oligostilbenoids have also been reported from the genera *Dipterocarpus* and *Upuna*, such as diptoindonesin E (**183**) from *Dipterocarpus hasseltii* [69], and upunaphenols C (**184**) and D (**185**) and *trans*-(**186**) and *cis*-upunaphenols K (**187**) from *Upuna borneensis* [96,34]. Table 4 summarizes the distribution of tetramer oligostilbenoids within Dipterocarpaceae. *Upuna, Vateria, and Vatica* exhibit varied types of tetramers, with 23, 22, and 19 reported compounds, respectively. In group A, *Vateria* is the primary source, followed by *Vatica* and *Hopea*. *Upuna*, *Vateria*, and *Vatica* serve as key sources in group B. Notably, (-)-hopeaphenol (**138**) is widely found across eight genera, followed by vaticanol B (**163**) in six genera and vaticanol C (**176**), hemsleyanols C (**167**), and D (**161**) each in four genera.

Table 4. Distribution of tetramer oligostilbenoids in the Dipterocarpaceae family.

No	Compounds	Sources
138	(-)-Hopeaphenol	*Balanocarpus heimii* and *Hopea odorata* [10], *H. parviflora* [39], *Shorea talura* [99], *S. robusta* [100], *S. hemsleyana* [42], *S. roxburghii* [101], *Neobalanocarpus heimii* [66], *Dipterocarpus grandiflorus* [49], *D. hasseltii* [69], *Vatica umbonata* [81], *V. albiramis* [34], *Vateria indica* [95], *Upuna borneensis* [96], *Dryobalanops lanceolata* [102], and *D. rappa* [72]
140	(+)-Isohopeaphenol	*Shorea hemsleyana* [42], *S. balangeran* [74], and *Vateria indica* [35]
141	Hopeaphenol A	*Vatica oblongifolia* [34]
142	Isohopeaphenol A	*V. oblongifolia* [34] and *Vateria indica* [95]
143	Pauciflorol C	*Vatica pauciflora* [53] and *Hopea parviflora* [39]
144	Vateriaphenol B	*Vateria indica* [35], *Vatica pauciflora* [53], and *Hopea parviflora* [39]
145	Vateriaphenol C	*Vateria indica* [95]
146	Neohopeaphenol A	*Hopea hainanensis* [67]
147	Neoisohopeaphenol A	*H. hainanensis* [67]
148	Vaterioside B	*Vateria indica* [32]
149	Vatalbinoside A	*Hopea parviflora* [39] and *Vatica albiramis* [34]
150	Diptoindonesin F	*Shorea gibbosa* [78]
151	Vateriaphenol D	*Vateria indica* [76]
152	Dibalanocarpol	*Hopea malibato* [59], *Vateria indica* [35], and *Shorea roxburghii* [77]
153	Stenophyllol A	*Upuna borneensis* [96], *Vateria indica* [32], and *Dryobalanops lanceolata* [102]
154	Pauciflorol B	*Vatica pauciflora* [53], *Upuna borneensis* [96], and *Vateria indica* [32]
155	Upunaphenol B	*Upuna borneensis* [96] and *Vateria indica* [76]

156	Upunaphenol I	*Upuna borneensis* [34]
157	Upunaphenol H	*Upuna borneensis* [34]
158	Upunaphenol P	*U. borneensis* [97]
159	Upunaphenol J	*U. borneensis* [34]
160	Hemsleyanol C	*Shorea hemsleyana* [61], *Vatica pauciflora* [53], and *Vateria indica* [32]
161	Hemsleyanol D	*S. gibbosa* [78], *Vatica pauciflora* [53], *Dipterocarpus grandiflorus* [49], and *Vateria indica* [32]
162	Vaticaphenol A	*Vatica diospyroides* [33], *V. oblongifolia* [34], and *Neobalanocarpus heimii* [66]
163	Vaticanol B	*Vatica rassak* [28], *V. pauciflora*[53], *Hopea utilis* [30], *H. dryobalanoides* [63], *Vateria indica* [35], *Dipterocarpus grandiflorus* [49], *Upuna borneensis* [82], *horea balangeran* [90], *Dryobalanops aromatica* [103], *D. lanceolata* [102], *D. rappa* [72], and *D. beccarii* [73]
164	Isovaticanol B	*Vatica pauciflora* [53] and *Upuna borneensis* [96]
165	Upunaphenol G	*U. borneensis* [41]
166	Vaticaside B	*Vatica rassak* [85], *V. indica* [35], *V. albiramis* [34], *Upuna borneensis* [54], and *Cotylelobium lanceolatum* [37]
167	Vaticaside C	*Vatica rassak* [85*]*, *V. albiramis* [34], *Upuna borneensis* [54], *Cotylelobium lanceolatum* [37], and *Vateria indica* [35]
168	Vatalbinoside B (**2**)	*Vatica albiramis* [34]
169	Upunaphenol F	*Upuna borneensis* [41] and *Vateria indica* [32]
170	Upunaphenol L	*Upuna borneensis* [98]
171	Upunaphenol M	*U. borneensis* [98]
172	Upunaphenol N	*U. borneensis* [98]
173	(-)-Ampelopsin H	*Shorea hemsleyana* [61] and *Vateria indica* [35]
174	Stenophyllol C/ Nepalensinol B	*Upuna borneensis* [96], *Vatica albiramis* [34], *Dryobalanops lanceolata* [102], and *D. rappa* [72]
175	Vatdiospyroidol	*V. diospyroides* [33]
176	Vaticanol C	*V. rassak* [28], *V. pauciflora* [53], *Dipterocarpus grandiflorus* [49], *Vateria indica* [35], *Upuna borneensis* [82], *Dryobalanops aromatica* [103], *D. lanceolata* [102], *D. rappa* [72], and *D. beccarii* [73]
177	Isovaticanol C	*Vatica pauciflora* [53]
178	Vaticanol F	*V. rassak* [85]
179	Grandiphenol A	*Dipterocarpus grandiflorus* [49], *Vateria indica* [32], and *Hopea parviflora* [39]
180	Grandiphenol B	*Dipterocarpus grandiflorus* [49]
181	Vateriaphenol F	*Vateria indica* [32]
182	Cotylelophenol C	*Cotylelobium lanceolatum* [37]
183	Diptoindonesin E	*Dipterocarpus hasseltii* [69]
184	Upunaphenol C	*U. borneensis* [96]
185	Upunaphenol D	*U. borneensis* [96], *Dryobalanops lanceolata* [102], and *D. rappa* [72]
186	*trans*-Upunaphenol K	*U. borneensis* [34]
187	*cis*-Upunaphenol K	*U. borneensis* [34]

Other Oligostilbenoids

In the Dipterocarpaceae family, only a few oligostilbenoids with a degree of polymerization greater than a tetramer could be found, with only 11 compounds reported, which include three pentamers, five hexamers, two heptamers, and one octamer. Most high-degree oligostilbenoids originate from the condensation of known dimer, trimer, or tetramer oligostilbenoids. Upunoside A (**188**), the first natural pentameric oligostilbenoid glucoside isolated from *Upuna borneensis* [54], shares identical stereochemistry of all chiral carbons with the tetramer hemsleyanol C (**160**), except for its stilbene glucoside unit, suggesting biogenetic formation from **160**. Additionally, hopeasides A (**189**) and B (**190**), both from *Hopea parviflora*, are stereoisomers with identical structures, as identified by Abe et al. [39], based on their optical rotation (Figure 7).

Five hexamer oligostilbenoids from the Dipterocarpaceae family are formed through the condensation of two trimers or a dimer and a tetramer. Upunaphenol A (**191**), isolated from *Upuna borneensis* [82], originates from the dimer ε-viniferin (**51**) and the tetramer vatdiospyroidol (**175**). Albiraminol A (**192**), obtained from *Vatica albiramis* [104], resulted from the dimer (-)-heimiol A (**88**) [104] and the tetramer vaticaphenol A (**162**). Hexamers from *Vatica rassak,* which include vaticanols D (**193**), I (**194**), and H (**195**) [105], are formed by condensing two trimer oligostilbenoids. Hexamers **193** and **195** are derived from the oligostilbenoid 'trimer A' [a degradation product of vaticanol G (**136**)] and the tetramer pauciflorols B (**113**), while hexamer **194** is produced from 'trimer A' and the tetramer vaticanol G (**136**) (Figure 7). Furthermore, two heptamer oligostilbenoids have been reported, which are vaticanol J (**196**) from *Vatica rassak* [105] and pauciflorol D (**197**) from *V. pauciflora* [60]. The structure of **196** resembles hexamers **193**, **194**, and **195**, and is derived from the condensation with the tetramer isovaticanol B (**164**). In contrast, **197**'s skeleton originates from the condensation of the trimer davidiol B (**132**) and the tetramer isovaticanol B (**164**). The largest oligostilbenoid identified to date is the octamer vateriaphenol A (**198**), discovered from the stem bark of *Vateria indica* [106,35] and later from *Vatica albiramis* [34]. The structure of **198** is formed from the condensation of two tetramer oligostilbenoids, (-)-hopeaphenol (**138**) and vaticanol B (**163**).

The distribution of oligostilbenoids with a degree of polymerization greater than that of a tetramer is very limited. The largest oligostilbenoids from Dipterocarpaceae have been reported in four genera: *Vatica, Vateria, Hopea*, and *Upuna*, with *Vatica* being the primary source of these oligostilbenoids. In another family, the largest oligostilbenoids were reported solely from the genera *Sophora* (Fabaceae) and *Vitis* (Vitaceae). Biogenetically, the condensation sites for the largest oligostilbenoids primarily occur at the aromatic carbons C12 or C3 with aliphatic carbons C7 or C8 (C12-C7/8 or C3-C7/8 types). Such types of condensations are uncommon in Dipterocarpaceae, which is why very few oligostilbenoids larger than a tetramer have been reported from this family.

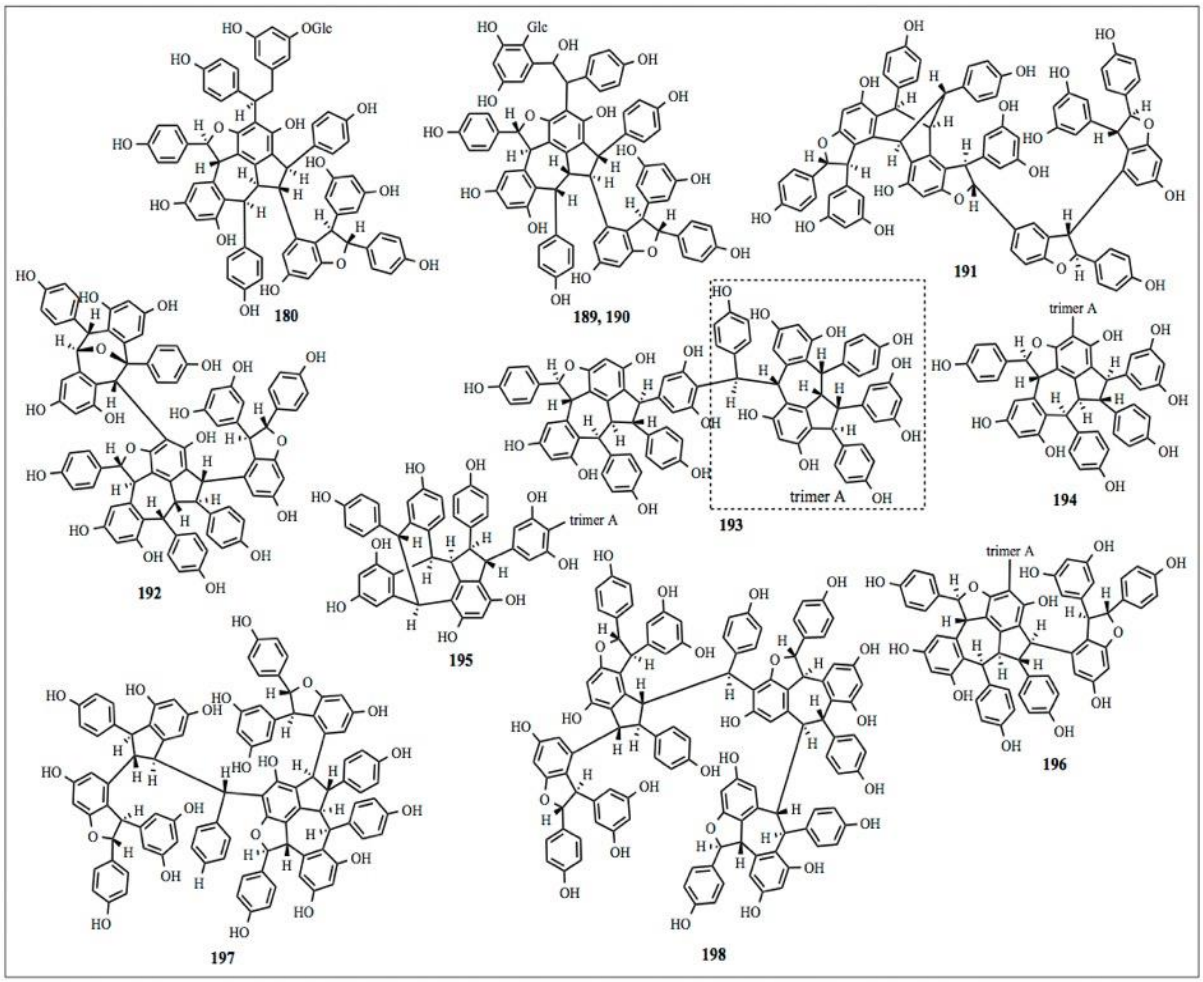

Figure 7. Structures of other oligostilbenoids from the Dipterocarpaceae family.

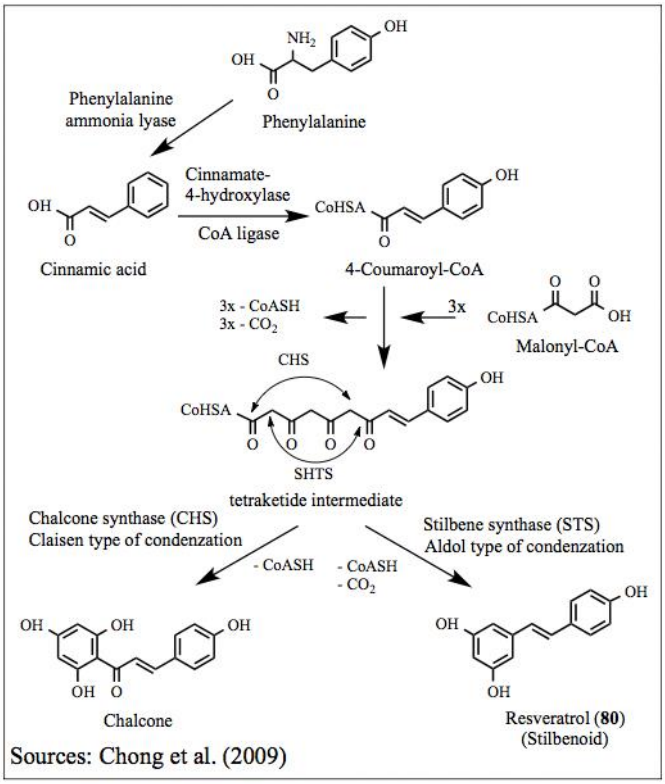

Figure 8. Biosynthesis pathways of stilbenoid and flavonoid.

Biogenesis of Oligostilbenoids

The biosynthesis of oligostilbenoids has not been extensively investigated. Yet, it is understood that oligostilbenes originate from monomeric resveratrol (**50**), with radical species serving as key intermediates in the biosynthesis of all oligostilbenoids. In recent years, the biosynthesis of simple stilbene derivatives has been well characterized [107,108], specifically requiring stilbene synthase (STS) for their synthesis [109,110]. Resveratrol (**50**) is biosynthesized from the combination of phenylalanine (via the shikimate pathway) and three acetate molecules derived from malonyl coenzyme A (acetate malonate pathway). Notably, the first step of this pathway is shared by both resveratrol and flavonoid biosynthesis; however, they diverge at the cyclization of a stilbene-3,5,7-triketoheptanoid acid (Figure 8). An aldol-type cyclization reaction of the polyketide intermediate results in the formation of stilbene-2-carboxylic acid, which is typically an unstable and bound intermediate in the pathway. Consequently, it is converted to resveratrol (**50**) or other derivatives. In a Claisen-type cyclization, the same polyketide intermediate reacts to produce a chalcone, leading to further modifications that yield various flavonoid derivatives [108]. Here, plant-specific polyketide synthases, such as stilbene synthase (STS) and chalcone synthase (CHS), play crucial roles in the biosynthesis of resveratrol and chalcones, respectively [109]. Although all higher plants can synthesize malonyl-CoA and cinnamic acid derivatives, only a few plant species can produce stilbene derivatives, as stilbene synthase is present in just a few plant families [110].

The resonance and delocalization of phenoxy anions or radicals through conjugation with unsaturated systems lead to a high degree of stabilization for these species within the benzene ring of compound **50**. Additionally, the olefinic carbons connecting the two aromatic rings in compound **50** show reactivity owing to the extended electron delocalization throughout the molecular framework (see Figure 9). These characteristics enhance compound **50**'s ability to participate in reactions as an ion or radical, particularly at its carbon-carbon center [111]. Notably, the polymerization of compound **50** typically initiates with the formation of carbon-carbon bonds through an oxidative coupling reaction involving its radical species. This is often followed by secondary reactions, such as the formation of a benzofuran ring or a bicyclo ring through ionic mechanisms.

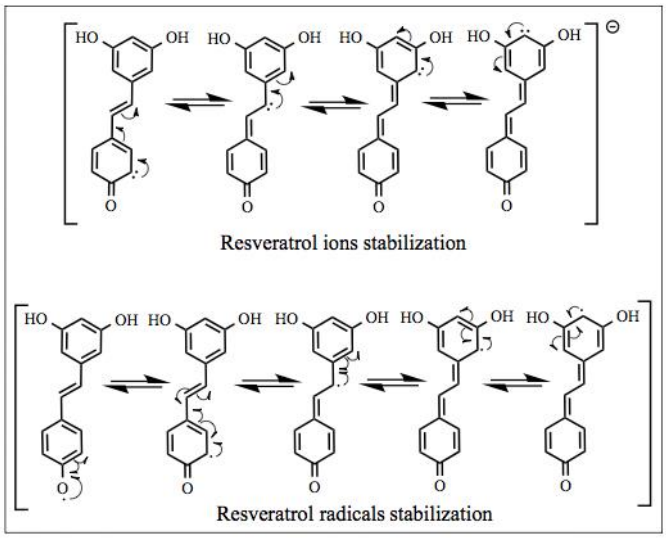

Figure 9. Resonance stabilization of phenoxy ions and radicals in resveratrol.

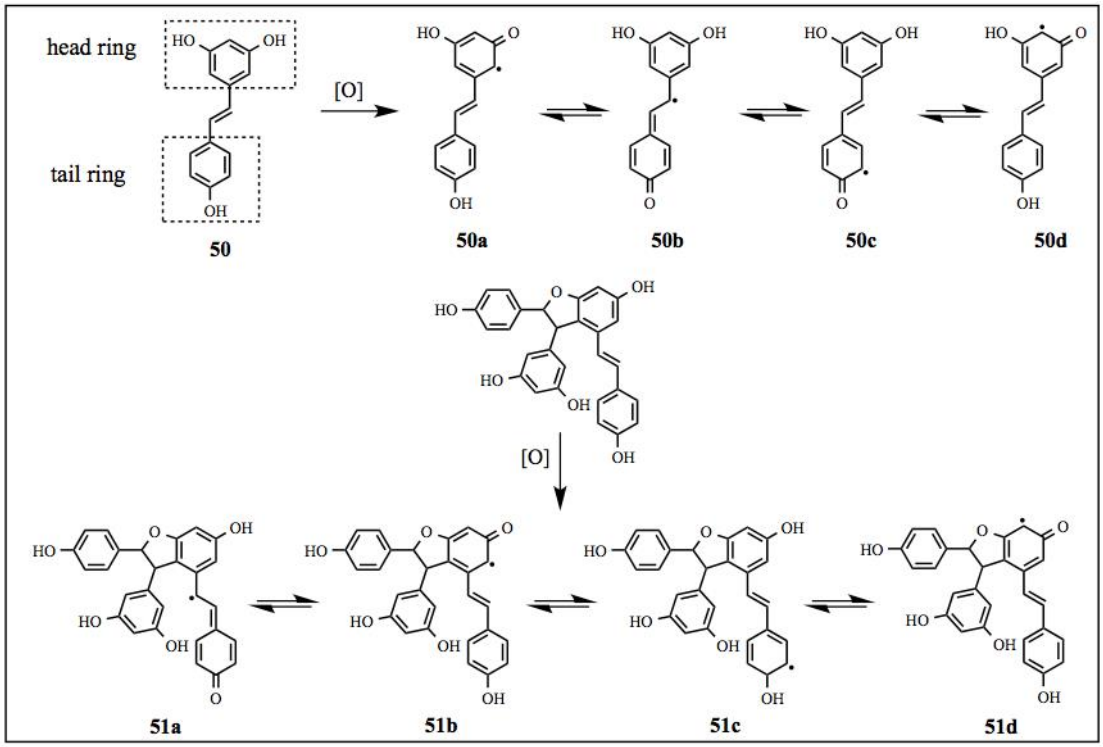

Figure 10. Important types of resveratrol (**50**) and ε-viniferin (**51**) radicals in the biosynthesis of oligostilbenoid.

In complex polymers, the monomer unit of **50** in the oligostilbenoid framework is recognized by two distinct ring structures: a 3,5-dihydroxybenzene ring, known as the head ring, and a p-hydroxybenzene ring, referred to as the tail ring. These rings are linked by ethylene or ethyl carbons (Figure 10). Structural modifications of **50**, including degradation or rearrangement reactions, are uncommon, simplifying the process of identifying this specific unit within the oligostilbenoid configuration. Furthermore, the oligomerization of **50** can be deduced from the resveratrol radicals **50a** to **50d**, which are the main precursors [111]. However, within the Dipterocarpaceae family, only radicals **50a** and **50b** are typically seen, while radicals **50c** and **50d** are more prevalent in other plant families. The presence of ε-viniferin radicals **51a** to **51d**, acting as advanced precursors, indicates oligostilbenoids forming with a high degree of polymerization. The introduction of precursor **51** into the oligostilbenoid structure is marked by a benzofuran ring. It is important to note that the benzofuran ring in oligostilbenoids may not solely originate from radical **51**, as it can also be produced during condensation reactions [14]. Like resveratrol (**50**) radicals, only radicals **51a** and **51b** are generally

found in dipterocarp plants, whereas radicals **51c** and **51d** are less frequently encountered.

In 1993, Sotheeswaran and Pasupathy [12] postulated that all oligostilbenes with a benzofuran ring derive from the dimer ε-viniferin (**51**), while those without it come directly from resveratrol (**50**). Researchers like Ito et al. [83], Yan et al. [92], and Takaya et al. [93] support this. The biomimetic synthesis [110-112] and total synthesis reactions [113-115] have confirmed these pathways, exemplified by compounds such as balanocarpol (**69**), stemonoporol (**134**), copalliferol A (**135**), and hopeaphenol (**138**). Figure 11 shows that the formation of ε-viniferin (**51**) begins with the oxidative coupling of resveratrol radicals **50a** and **50b**, followed by intermolecular cyclization. Other oligostilbenoids will derive from dimer **51**, such as **69** which is formed from **51** through the cyclization of its olefinic and aromatic carbons, followed by oxidation. Meanwhile, compound **138** is produced from the condensation of two molecules of the radical intermediate **69a**. The formation of a seven-membered ring in **138**, occurring either before or after the tetramerization of **51**, as well as the hydroxylation of **69** following the formation of the seven-membered ring, is currently debated. Additionally, oligostilbenoids without benzofuran ring, such as stemonoporol (**134**) and copalliferol A (**135**), are derived directly from the condensation of three molecules of **50**, resulting from the direct condensation and cyclization of these radicals.

In the formation of **138**, Yan et al. [92] and Takaya et al. [93] proposed a different route that does not involve **69a** as an intermediary. In their approach, two molecules of radical **51a** condense via an intermediate **138a**, followed by intermolecular cyclization to yield a seven-membered ring (Figure 12). The formation of **69** through the hydroxylation reaction of intermediate **69a** using radical OH• remains a contentious topic, as it is well established that the hydroxylation of natural compounds occurs in the presence of O_2 alongside NADPH as a source of the hydroxyl group via an epoxidation reaction [116]. The use of radical OH• as a hydroxylation agent is uncommon because it is a highly reactive species that can damage enzymes or lipids through oxidation [117]. Therefore, it can be concluded that radical **69a** does not exist, and the formation of tetramer **138** is suggested to begin from the condensation of two molecules of radical **51a**, as illustrated in Figure 12.

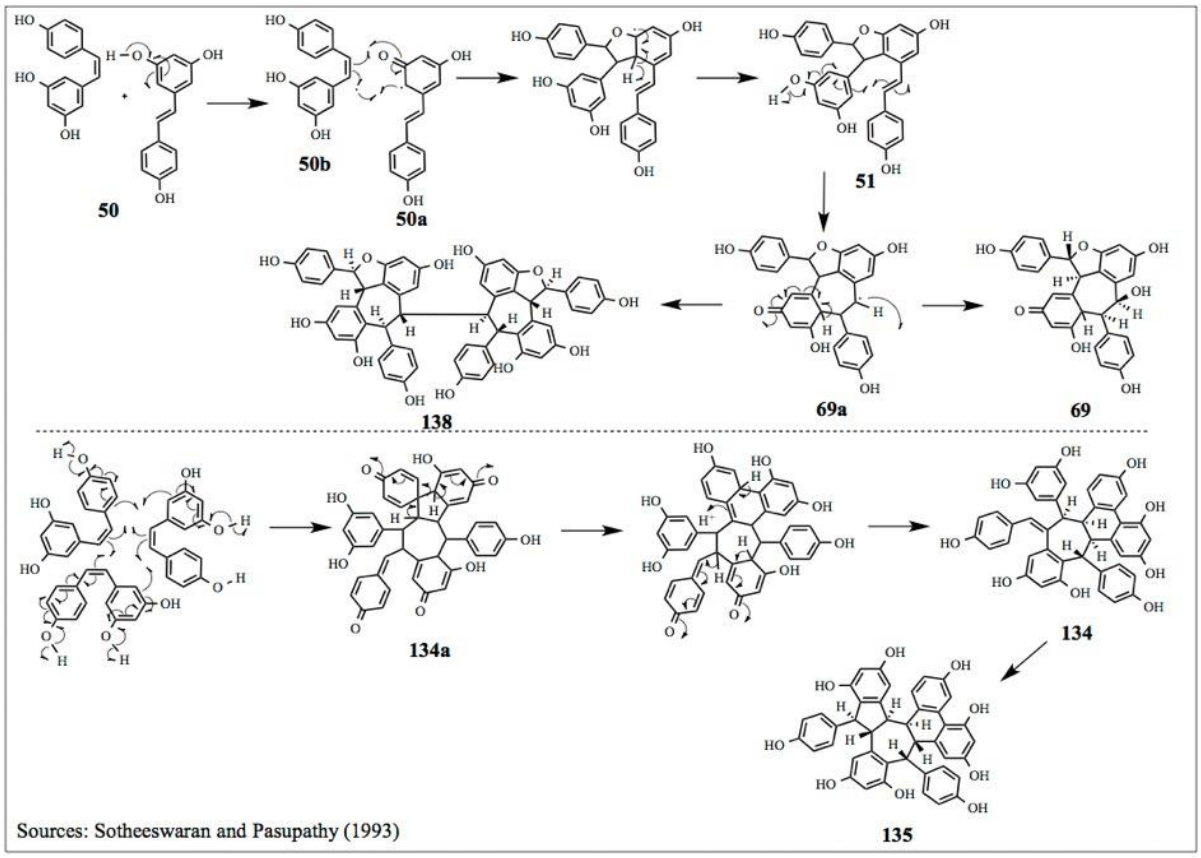

Figure 11. Plausible biogenetic pathways of several oligostilbenoids.

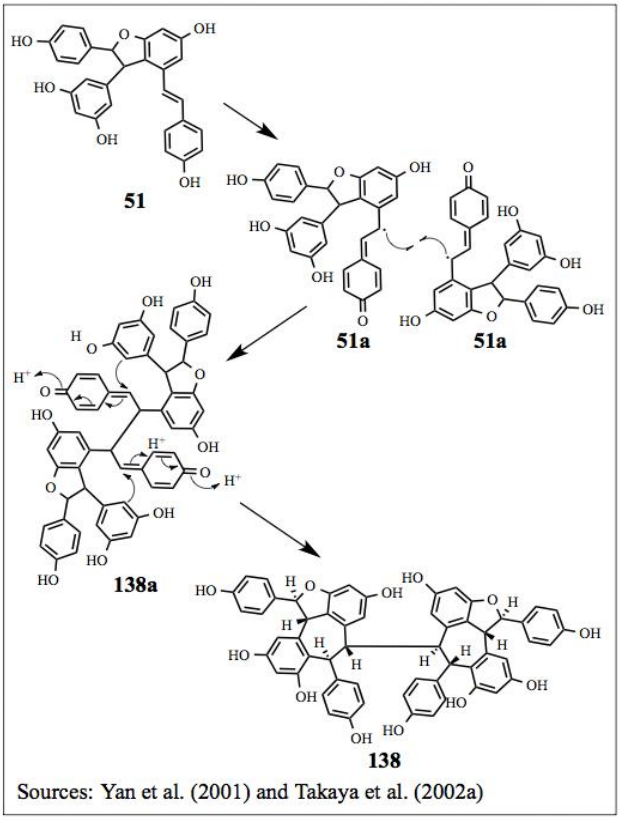

Figure 12. Plausible biogenetic pathways of hopeaphenol (**138**) from dimer ε-viniferin.

Takaya et al. [118] suggested that ε-viniferin (**51**) also plays an important biogenetic role in the formation of ampelopsins D (**90**) and F (**93**), which do not contain benzofuran rings. According to Takaya et al., the process begins with the protonation of the oxygen atom in the benzofuran ring. This is followed by a nucleophilic attack on the double bond, resulting in the formation of a five-membered ring in intermediate **51a**. The enolization of this intermediate then yields the skeleton of **90** (route a), while intermolecular cyclization of intermediate **51a**, followed by enolization, produces the skeleton of **93** (route b) (Figure 13). However, Velu et al. [113], Snyder [114], and Cai and Cai [,119] demonstrated a biomimetic synthetic approach demonstrating that both compounds can be synthesized from the condensation of two resveratrol analogue molecules. They concluded that the synthesis of **90** and **93**, both absent of a benzofuran ring, is achievable smoothly and selectively, aligning with Sotheeswaran and Pasupathy [12].

Bioactivities

The bioactivities of several secondary metabolites from the Dipterocarpaceae, such as oligostilbenoids, sesquiterpenes, triterpenes, and phenolic compounds, have been reported widely [13,14,120-122]. However, many researchers have shown greater interest in studying the oligostilbenoid constituents, as this is the most distinctive group within this family. Numerous studies have indicated that this group exhibits potent biological activities, including antimicrobial, antioxidant, anticancer, hepatoprotective, neuroprotective, anti-inflammatory, antidiabetic, and many more. Various *in vitro* assays on oligostilbenoids have been reported, however *in vivo* assays have been less documented due to the small quantities of oligostilbenoids that can be isolated from the plants. Additionally, the biological activities of other secondary metabolites, such as sesquiterpenes and triterpenes from this family, have also been reported' however all assays on these groups were based on constituents from plant families other than the Dipterocarpaceae.

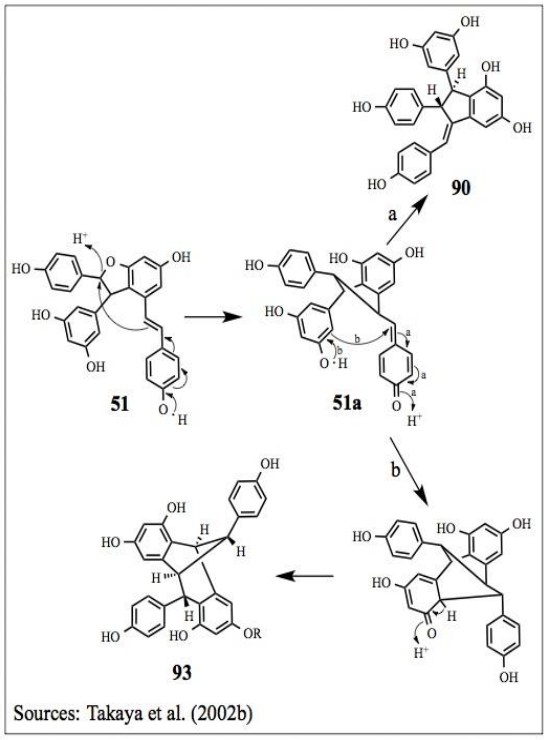

Figure 13. Plausible biogenetic pathways of ampelopsins D (**90**) and F (**93**) from the dimer ε-viniferin.

Antimicrobial Activity

The question of whether resveratrol qualifies as a true phytoalexin has remained unclear for some time. Kuc [123] defines phytoalexins as antimicrobial compounds that plants synthesize and accumulate in response to abiotic or biotic elicitors. They play a crucial role in the active defense mechanisms of plants. According to their unit structures, Wibowo et al. [72] conducted a detailed study on the structural features that influence the antibacterial activity of certain oligostilbenoids. Their findings highlight that the presence and number of benzofuran rings, as well as the availability of a free stilbene skeleton, are key determinants in enhancing antibacterial efficacy. These structural features are believed to contribute to bioactivity by facilitating interactions with bacterial targets, potentially disrupting essential cellular processes. On the other hand, their research suggests that other structural factors, including the presence of a carbonyl functional group, a symmetrical molecular framework, the total number of hydroxyl groups, and the overall degree of polymerization, do not have a significant impact on antibacterial potency. This implies that while these features may influence other chemical properties, they do not directly contribute to the mechanism responsible for antibacterial effects.

In 1977, Langcake and Pryce [46] demonstrated the antifungal activities of ε- (**51**) and α-viniferin (**98**) against *Cladospohum cladosporioides*, *Botrytis cinerea*, *Plasmopara viticola*, and *Piricularia oryzae*. Canaliculatol (**126**) was only active against *Cladospohum cladosporioides* [87]. Using the NCCLS protocol M38-P, resveratrol (**50**) showed activities against several fungi, including *Trichophyton mentagrophytes*, *T. tonsurans*, *T. rubrum*, *Epidermophyton floccosum*, and *Microsporum gypseum* (LD_{50} 25–50 μg/ml) [124], as well as *Botrytis cinerea* (LD_{50} 60 μg/ml; 2.6×10^{-4} M) [125]. Its glucoside derivative, trans-piceid (**52**), significantly inhibited the germination of *Venturia inaequalis* (200-400 μg/ml). Additionally, Ge et al. [31] reported the antifungal activities of seven oligostilbenoid derivatives against other plant pathogens, including *Alternaria altenata*, *A. solani*, *Colletotrichum lagenarium, Fusarium oxysporum*, *Pyricularia oryzae*, and *Valsa mali*. Of these, only hopeanolin (**100**) demonstrated activities against six types of pathogenic fungi with MIC values ranging from 0.10 to 22.5 μg/ml, while the other compounds were effective against specific fungi.

In antibacterial assays, using the paper disc method, ε-viniferin (**51**) and miyabenol C (**104**) exhibited moderate activities against *Staphylococcus aureus* [126, 127]. However, Yim et al. [128] reported that ε-viniferin displayed stronger activities against *S. mutans* and *S. sanguis*, with MIC values of 25 and 12.5 μg/ml, respectively. Nitta et al. [129] and Abe et al. [34] found that oligostilbenoid derivatives, including α-viniferin (**98**), vaticanol B (**163**), hopeaphenol (**138**), and hemsleyanol D (**161**), showed antimicrobial activities against methicillin-resistant *S. aureus* (MRSA), with **161** being the most effective (MIC of 2 μg/ml). Other oligostilbenoid derivatives, such as distichol (**125**) and copalliferols A (**135**) and B (**133**), demonstrated only moderate activities against *Oxford*

staphylococcus and *Escherichia coli* [89,47,91,88,48]. Non-oligomeric compounds, such as caryophyllene (**3**), exhibited antibacterial activities against *Staphylococcus mutans* and *Propionibacterium acnes* [130]. Ursolic acid (**16**) was active against *Mycobacterium tuberculosis* H37Rv, 99% inhibition at 100 μg/ml [131]. Additionally, p-coumaric acid (**43**) inhibited the type III secretion system (T3SS), a crucial virulence factor in many Gram-negative pathogens [132]. Furthermore, other antimicrobial activities of compounds from Dipterocarpaceae plants, such as anti-babesial and anti-HIV effects, have also been reported. For example, vaticanol A (**111**) inhibited the growth of *Babesia gibsoni in vitro* at a concentration of 25 μg/mL [90], and balanocarpol (**70**) was tested in the NCI primary anti-HIV screen *in vitro,* showing a very modest HIV-inhibitory activity (EC_{50} values of 20 μg/mL) [59]. However, resveratrol (**50**) possessed only an anti-HIV-1 PR activity, with an IC_{50} value of 85.0 μg/mL [133].

Antioxidant Activity

Enzymatic and nonenzymatic processes in cells always generate various reactive oxygen species (ROS). If excessive ROS are not promptly eliminated from the cells, the accumulated ROS can induce cytotoxicity, which plays a major role in the pathogenesis of a wide variety of diseases, including cardiovascular issues, cancer, diabetes, Alzheimer's disease, and autoimmune disorders. Antioxidants can disrupt the initiation and propagation steps of the ROS chain reaction, playing an important protective role against the damage caused by various diseases.

Resveratrol (**50**) is renowned for its antioxidant activity [134], demonstrating 89.1% inhibition of lipid peroxidation in a linoleic acid emulsion at 30 μg/ml [135], surpassing BHA, BHT, α-tocopherol, and trolox, which showed inhibition values of 83.3%, 82.1%, 68.1%, and 81.3%, respectively. Its glucoside derivatives, trans-piceid (**52**) and resveratrol-3-C-β-glucopyranoside (**57**), also exhibit antioxidant properties, inhibiting lipid peroxidation and xanthine oxidase *in vitro* [136, 137]. Additionally, ε-viniferin (**51**) demonstrates the highest antioxidant capacity in the DMSO/O_2-polar system (IC_{50} 0.14 mM) [138], significantly inhibiting the degradation of 2-deoxyribose and lipid peroxidation in rat liver microsomes (IC_{50} 0.17 mM and 0.41 mM, respectively) [139]. Other oligostilbenoids, malibatol A (**74**) and hopeahainol C (**111**), displayed potent antioxidant and radical scavenger abilities at 0.4 mM compared to resveratrol, ascorbic acid, and BHA [38]. Lastly, vaticanol D (**193**) showed an activity against superoxide in the xanthine-xanthine oxidase system (IC_{50} 7.4 mM) [140].

Anti-inflammatory Activity

Inflammation is characterized by the release of pro-inflammatory cytokines, including tumor necrosis factor-α (TNF-α), interleukin-1β (IL-1β), and interleukin-6 (IL-6). Additionally, inflammatory mediators such as nitric oxide (NO) and prostaglandin E2 (PGE2) are synthesized by inducible nitric oxide synthase (iNOS) and cyclooxygenase (COX) [141, 142]. These cytokines and mediators contribute to the development of several human diseases, including rheumatoid arthritis, asthma, atherosclerosis, and endotoxin-induced multiple organ injury [143,144]. In these cases, anti-inflammatory agents can help reduce the inflammatory response by decreasing the production of these inflammatory cytokines and mediators [145, 146].

The anti-inflammatory activity of oligostilbenoids is primarily influenced by their hydroxyl (OH) groups, the stilbene backbone, and specific structural modifications that enhance interactions with inflammatory mediators. Hydroxyl groups, particularly at the C3, C5, and C4' positions, contribute to radical scavenging and inhibition of pro-inflammatory enzymes such as COX-2 and 5-LOX through hydrogen bonding [147]. The trans-stilbene backbone facilitates π-π stacking interactions with target proteins, stabilizing anti-inflammatory pathways. Methoxy (OCH_3) substituents may enhance lipophilicity and improve cell membrane permeability, aiding intracellular bioavailability [148]. Additionally, oligomerization influences potency, as dimeric and trimeric forms exhibit enhanced binding to inflammatory cytokines like TNF-α and IL-6, thereby reducing inflammatory responses [149]. The combination of hydroxylation, conjugated double bonds, and structural flexibility plays a crucial role in modulating anti-inflammatory activity.

Resveratrol (**50**) is recognized as a powerful anti-inflammatory agent and continues to garner interest. Research has shown that resveratrol (**50**) suppresses the expression of pro-inflammatory markers such as TNF-α, IL-6, IL-8, and COX-2 by decreasing intracellular levels of Ca^{2+} and ERK 1/2, while also activating NF-κB in the human mast cell line (HMC-1) [150]. Other studies indicated that resveratrol (**50**) leads to significant reductions in levels of pro-inflammatory cytokines TNF-α and IL-1β and an increase in the anti-inflammatory cytokine IL-10. It also reduces the expression of PGES-1, COX-2, and iNOS proteins by downregulating p38, a mitogen-activated protein kinase (MAPK) signaling pathway in resveratrol-fed animals [151]. Additionally, Liu et al. [152] reported that vaticanol B (**163**) and vaticanol C (**176**) can inhibit IgE-mediated histamine release and reduce TNF-α and leukotriene productions triggered by IgE in bone marrow-derived mast cells (BMMC). Meanwhile, α-viniferin (**98**) and vaticanol C (**176**) were shown to strongly inhibit the activation of extracellular signal-regulated kinases (ERK) mediated by IgE. Furthermore, the impact of certain oligostilbenoid derivatives on the interleukin-1β-induced production of matrix metalloproteinase-1 (MMP-1) in human dermal fibroblasts was examined. Hopeaphenol (**138**) and vaticanol C (**176**) were found to significantly inhibit MMP-1 production [34].

Anticancer Activity

The anticancer effects of oligostilbenoids typically involve inducing cytotoxicity, inhibiting proliferation, or triggering apoptosis in cancer cells. A previous study found that resveratrol (**50**) exhibited significant cytotoxicity against various human cancer cell lines, including liver hepatoma HepG2 (IC_{50} 11.8), colon HT-29 (IC_{50} 25.2 μg/ml) [153], colon SW480 (IC_{50} 22.1 μM), and promyelocytic leukemia HL-60 (IC_{50} 13.1 μM) [154]. Furthermore, Ito et al. [154] reported the cytotoxicity of several compounds, including ε-viniferin (**59**), vaticaside B (**166**), vaticanols B (**163**), C (**176**), and D (**193**), hemsleyanol D (**161**), and vateriaphenol A (**198**) against the same cancer cell lines, SW480 and HL-60. The assays showed that compounds **59**, **176**, **193**, **161**, and **198** were active against both cancer cell lines, with IC_{50} values of 18.5/5.2, 3.2/3.0, 8.9/9.8, 32.5/12.3, and 9.8/9.7 μM, respectively. On the other hand, compounds **166** and **163** were only active against the HL-60 cell line, with IC_{50} values of 5.1 and 4.8 μM, respectively. In other assays, trans-piceid (**52**) demonstrated activities against mouse leukemia L1210 (IC_{50} 28.7 μM) and human leukemia K562 (IC_{50} 24.6 μM) [155]. Additionally, α-viniferin (**98**) showed activities against human colon HCT-116, HT-29, and Caco-2 cell lines (IC_{50} 6-32 μM) [156]. Hopeaphenol (**138**) was significantly active against the human epidermoid carcinoma KB cell line (ED_{50} 1.2 μg/ml) [157]. Vatdiospyroidol (**175**) exhibited activities against KB (EC_{50} 1.0 μg/ml), Col2 (EC_{50} 1.9 μg/ml), and BC1 (EC_{50} 3.8 μg/ml) cell lines [33]. Finally, vaticanol D (**193**) and vateriaphenol A (**198**) showed activities against the KB cell line with ED_{50} values of 11.8 μM [105] and 10.5 μM [106], respectively.

On the other hand, several studies also reported that compound **50** possesses a potential as a cancer chemopreventive agent, exhibiting effects on the initiation, promotion, and progression of cancer cell growth [121]. *In vitro* assays demonstrated that compound **50** prevented the initiation of tumor formation through antimutagenic effects, induced the carcinogen-detoxifying phase II enzyme quinone reductase [158], and inhibited the expression of the phase I enzyme CYP1A1 [159], which is responsible for procarcinogen activation. Additionally, a study by Kang et al. [160] indicated that compounds **50** and **51** reduce the viability of HL-60 cells in a dose-dependent manner (IC_{50} 20-90 μM), significantly suppressing HL-60 cell proliferation and inducing DNA damage. Balanocarpol (**69**) and ampelopsin A (**71**) were shown to inhibit sphingosine kinase-1 activity, resulting in down-regulation of its expression, reduced DNA synthesis, and stimulation of PARP cleavage in the MCF-7 breast cancer cell line [161]. Other oligostilbenoids, such as miyabenol C (**104**), induced apoptosis in the myeloma cell line via mechanisms entirely dependent on caspase activation, which is associated with disruption of mitochondrial membrane potential [162]. Additionally, nepalensinol B (**174**) exhibited a potent inhibitory effect on DNA topoisomerase II (IC_{50} 0.02 μg/ml) [163]. At the same time, vaticanol C (**176**) inhibited the phosphorylation of both ERK and Akt in the HL-60 cell line, leading to reduced phosphorylation of Bad [164].

Other Activities

In an antidiabetic assay, Morikawa et al. [165] reported that hopeaphenol (**138**) and isohopeaphenol (**139**) exhibited inhibitory effects on plasma triglyceride elevation at a dose of 200 mg/kg body weight. These compounds demonstrated pancreatic lipase inhibitory activities with IC_{50} values of 32.9 μM and 26.5 μM, respectively. Additionally, in another study, Morikawa et al. [77] reported that α-viniferin (**97**), balanocarpol (**69**), malibatols A (**74**) and B (**75**), hopeafuran (**77**), vaticanol C (**176**), hemsleyanol D (**161**), and vaticaside B (**166**) showed inhibitory activities against plasma glucose elevation in sucrose-loaded rats at doses ranging from 100 to 200 mg/kg body weight. This group of compounds also inhibited rat lens aldose reductase with IC_{50} values of 7.8, 30.0, 35.6, 10.0, 6.9, 21.2, 29.4, and 30.0 μg/ml, respectively. Furthermore, ε-viniferin (**51**) and vaticanol C (**176**) were active against PPARα and PPARβ/δ, which are nuclear receptor proteins that act as transcription factors. These receptors regulate gene expression and play crucial roles in carbohydrate metabolism regulation [166].

Recent studies by Zghonda et al. [167] have demonstrated the effects of resveratrol (**50**) and (-)-ε-viniferin (**51**) on the functions of vascular endothelial cells (VECs), as well as on blood pressure and cardiac mass in spontaneously hypertensive rats (SHRs). These studies suggest that both compounds may play a role in protecting VECs and maintaining heart function. Additionally, (+)-ε-viniferin (**61**) exhibited antiangiotensin-converting enzyme (anti-ACE) properties and vasodilating effects in an endothelium-intact aortic ring in rats [168], specifically against phenylephrine-induced tensions. Furthermore, α-viniferin (**97**) significantly inhibited acetylcholinesterase (AChE) activity in a dose-dependent manner with an IC_{50} of 2.0 μM [169]. In another study, the antihepatotoxic activity of ampelopsin E (**103**) was reported using a cytotoxicity model system with primary culture rat hepatocytes. At a dose of 0.1 μg/ml, compound **103** reduced serum GPT levels by 64% compared to the control, demonstrating its protective effect against carbon tetrachloride-induced liver damage in rat hepatocytes [170].

CONCLUSION

The Dipterocarpaceae family is a valuable source of diverse secondary metabolites, particularly oligostilbenoids, which hold promise for ecological studies and medicinal applications. These compounds exhibit significant bioactivities, including antimicrobial, anticancer, and anti-inflammatory properties, underscoring their pharmaceutical potential. Variations

in metabolite profiles across genera aid in taxonomic studies and the discovery of novel bioactive compounds. While progress has been made in understanding these metabolites' chemical and biological properties, further research is essential to explore their therapeutic potential and address challenges in sustainable extraction and application challenges. Future studies should focus on these compounds' synthesis, bioavailability, and clinical efficacy to fully realize their promise in modern medicine.

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