REVIEW ARTICLE

Genus *Prismatomeris* (Rubiaceae): Phytochemistry and their biological activities

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Abstract

Prismatomeris is a genus of the family Rubiaceae, which is mainly distributed in Southeast Asia. Some of the species were used to treat wounds, hepatitis, anemia, leukocythemia, pneumoconiosis, bronchitis, and as aphrodisiac. Up to now, the reported constituents from the genus *Prismatomeris* include anthraquinones, iridoids, and triterpenoids. Among them, anthraquinones, the characteristic components of this species become an intention due to their complex structures and significant biological features such as cytotoxic, antitumor, anticancer, antifungal, antimalarial, antiplasmodial, and antituberculosis activities. This review presents a systematic compilation of available data of secondary metabolites and their bioactivity studies of the genus *Prismatomeris*. It may lead to upcoming drug design, and therefore, provide a reference for advanced study and application of *Prismatomeris*.

Key words: Anthraquinones, cytotoxicity, iridoids, pharmacology, *Prismatomeris,* Rubiaceae

INTRODUCTION

The genus *Prismatomeris* (Rubiaceae) comprises about 25 species, being distributed in the tropical and subtropical areas in Southeast Asia. It is classified in the tribe of *Morindeae*, together with *Morinda* and *Renellia*; it seems closely allied to the latter genus. However, some researchers have classified it in a separate tribe; *Prismatomerideae*.[1] The genus *Prismatomeris* is a small shrub tree, with glossy pale brown bark flaking off when dry, and each internode with a median longitudinal ridge ending between each pair of leaf stalks. The leaves are ovate to elliptic or slightly obovate. Secondary veins are known to inarch to form a series of marginal loops while tertiary veins are reticulate or obscure. The stipules of the leaves are triangular, bifid at the tip and the apices are either look like two cusps or fine points. Its flowers are stalked or sessile, in terminal and axillary clusters of two to ten bisexual and heterostylous. The calyx cup fringed by four to six teeth or subtruncate. The color is white. The ovary is two-celled while ovules one per cell. It attached to ovary cross wall with filiform style and two-lobed stigma. The fruit is globose to ellipsoid with one or two seeded. The seeds are globose, with a narrow lateral groove at its point of attachment.[2-4] Previous phytochemical investigation of this genus was found to have anthraquinones, anthraquinone glycosides, iridoids, and triterpenoids.[5-13] The iridoids and triterpenoids are classified under the terpene groups while the anthraquinones are considered as phenolics. These compounds have shown cytotoxic, antitumor, anticancer, antifungal, antimalarial, antiplasmodial, and antituberculosis activities.[9,13,14,15]

The intention of this review is to study from phytochemical and bioactivity perspectives of the *Prismatomeris* genus for which the extraction, isolation, structural elucidation, and explanation of the bioactivities of compounds reported

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in literature. A substructure search performed using the SciFinder Scholar database and searched using the keyword *Prismatomeris* in PubMed, Medline, Scopus, Google Scholar, EBSCO, Directory of open access journals, EMBASE, and Web of Science. It was indicated that to date, six species have been cited in this perspective. For each plant, a discussion on its phytochemistry and their bioactivities is provided.

TRADITIONAL USES

In the traditional folklore medicine, *Prismatomeris* genus has been reported for the treatment of several ailments. In Peninsular Malaysia and Thailand traditional medicinal system, the leaves have been applied as a poultice to fresh wounds. The root of *Prismatomeris connata* has been used in traditional medicine in China for the treatment of hepatitis, anemia, leukocythemia, and pneumoconiosis.[2] The water decoction of the roots of *Prismatomeris fragrans* is used traditionally as a tonic.[3] In Thailand, the macerated roots in water are used to treat snakebites whereas, in Indochina, the decoction has been used in a mixture with coconut and henna to treat bronchitis.[4] In Malaysia, the aqueous extracts of the roots of *Prismatomeris glabra* have been used traditionally for wellness, enhancing stamina, and for its ergogenic effects.[16] In addition, as stated by a local taxonomist, this plant has been used as an aphrodisiac. The roots of *Prismatomeris tetrandra* were used as Chinese traditional medicine to treat leukocythemia, gum bleeding, hepatitis, and anemia.[17]

PHYTOCHEMISTRY

Since the 1980s, compounds 1–67 [Figure 1] were successfully isolated from *Prismatomeris* genus. Their structures, names, and the corresponding plant sources are discussed below. A literature survey revealed that only six species of *Prismatomeris* genus have been investigated worldwide which are, *Prismatomeris connate*,[7,8,12,13,18] *P. fragrans*,[15] *P. glabra*,[11,19,20] *Prismatomeris malayana*,[9] *Prismatomeris sessiliflora*,[5] and *Prismatomeris tetrandra*.[6,10,18,20-22] Chemical investigations have been reporting the existence of anthraquinones, iridoids, phenols, triterpenoids, and phytosterols.

P. connata Y. Z. Ruan

P. connata is distributed in Hainan, China where the root is also known by the common name *huang-gen* and has been used in traditional herbal medicine.[4] Pharmacological studies showed that the roots possess antibacterial, anti-inflammatory, and antitumor activities.[23] The first phytochemical study of this plant appeared in literature in 2011 when Hao *et al*. isolated from the root extracts and structurally characterized six known anthraquinone glycosides. They were 1-*O*-methylrubiadin 3-*O*-β-primeveroside **1**, damnacanthol 3-*O*-β-primeveroside **2**, rubiadin 3-*O*-β-primeveroside **3**, lucidin 3-*O*-β-primeveroside **4**, 1,3-dihydroxy-2-(methoxymethyl) anthraquinone 3-*O*-β-primeveroside **5**, and digiferruginol ω-gentiobiose **6**. Hydrolisis of **4** has afforded lucidin **7**.[13] In the same year, Feng *et al*. obtained a new anthraquinone from root extracts; 4-hydroxy-1,2,3-trimethoxy-6-methylanthracene-9,10-dione **8**, and two novel tetrahydroanthraquinones; prisconnatanones A **9** and B **10**, together with 15 known anthraquinones which are, 1-hydroxy-2,3-dimethoxy-7-methyl-9,10-anthraquinone **11**, 1,3-dihydroxy-2-methyl-anthraquinone **12**, ibericin **13**, lucidin ω-methyl ether **14**, 1,3-dihydroxy-5,6-dimethoxy-2-methyl-9,10-anthraquinone **15**, 3-hydroxy-1,5,6-trimethoxy-2-methyl-9,10-anthraquinone **16**, 3-hydroxy-1-methoxy-2-methyl-9,10-anthraquinone **17**, 2-methylanthraquinone **18**, lucidin**7**, 2-methoxyanthraquinone **19**, 1-methoxy-2-methylanthraquinone **20**, 2-hydroxy-1-methoxyanth raquinone **21**, 1,2,3-trimethoxy-7-methylanthr aquinone **22**, 1,3-dihydroxy-5,6-dimethoxy-2-(methoxymethyl)-9,10-anthraquinone **23**, and 6-methoxyibericin **24**.[7] A year later, a further reinvestigation by the same researchers also of the root extracts has afforded a new phenolic glycoside, prismaconnatoside A **25**, together with one phenolic glycoside, berchemolide **26** and four aspruloside type iridoid glycosides, aspruloside **27**, asperulosidic acid **28**, deacetylasperuloside **29**, and deacetylasperulosidic acid **30**.[18] 3 years later, Wang *et al*. managed to isolate seven new tetrahydroanthraquinones, prisconnatanones C–I **31-37** from the ethanol root extracts of *P. connata*. These new natural compounds belonged to the rare tetrahydroanthraquinone structural class.[12]

P. fragrans E. T. Geddes

*P. fragrans*is a tree of 2–12 m in height found in the Northeastern, Eastern, and Southeastern parts of Thailand and the Northwest of Laos.[1] It is known as *khao-san* in Nakhon Phanom Province.[3] The hexane and CH_2Cl_2 extracts of air-dried roots and stems of *P. fragrans* have been investigated by Kanokmedhakul *et al*.[15] They managed to isolate a new 1,3-dihydroxy-2-methyl-5,6-dimethoxyanthraquinone **38**, six known anthraquinones; nordamnacanthal **39**, damnacanthal **40**, rubiadin **12**, rubiadin-1-methyl ether **17**, lucidin-ω-methyl ether **14**, and 1-hydroxy-2-hydroxymethyl-3-methoxyanthraquinone **41**; β-sitosterol42; together with two known triterpenoids; 3β-acetylolean-12-en-28-olic acid **43**, and 3β-*O*-acetyl-11α,12α-epoxyolean-28,13-olide **44**. Modification of compound **38** furnished the methyl ether derivatives **45**.

P. malayana Ridley

*P. malayana*is a small tree up to 7 m tall and found indigenous to Western Malaysia. At the beginning of the 1960s, Lee described the isolation from root extracts and successfully identified rubiadin **12** and rubiadin-1-methyl ether **17**.[24] Tuntiwachwuttikul *et al*. continued the investigation on this species and successfully isolated a novel anthraquinone,

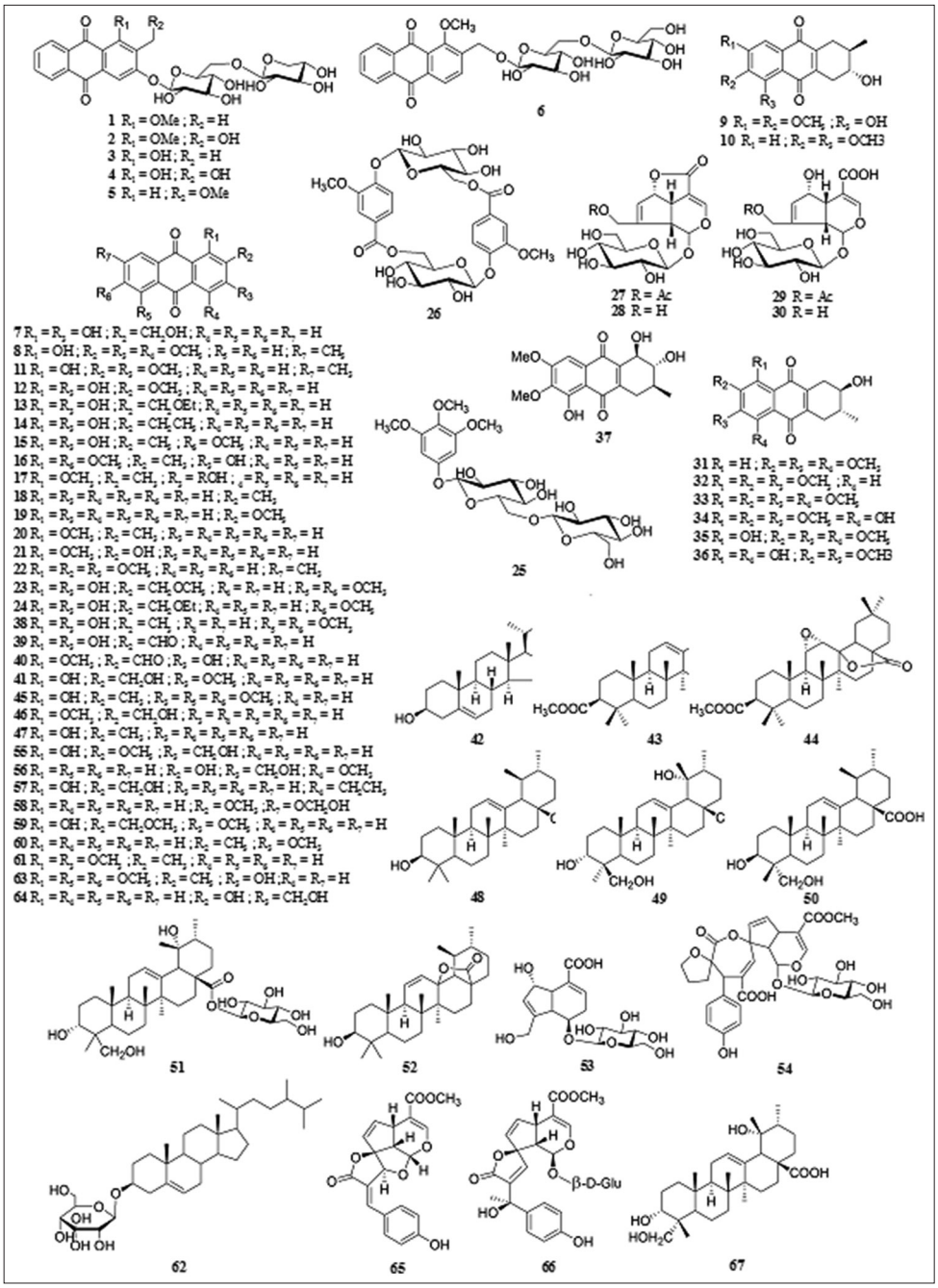

Figure 1: Chemical structures of the compounds isolated from the genus *Prismatomeris*

1,3-dihydroxy-5,6-dimethoxy-2-methoxymethyl-9,10-anthraquinone **23** and a compound, 2-hydroxymethyl-1-methoxy-9,10-anthraquinone **46**, along with seven known anthraquinones; tectoquinone **18**, 1-hydr oxy-2-methyl-9,10-anthraquinone **47**, rubiadin **12**, rubiadin-1-methylether **17**, 1,3-dihydroxy-5,6-dimethoxy-2-methyl-9,10-anthraquinone **38**, nordamnacanthal **39**, and damnacanthal **40**.[9] Recently, Abdullah has investigated the phytochemical studies from the leaves, roots, and stems of *P. malayana.* The isolation work on the leaves, roots, and stems yielded ursolic acid **48**, barbinervic acid **49**, 3β,23-dihydroxyurs-12-en-28-oic acid **50**, 28-*O*-β-glucopyranosyl-3α,19α,23-trihydroxyurs-12-en-28-oic acid **51**, 3β-hydroxyurs-11-en-13,28-olide **52**, asperulosidic acid **27**, scandoside **53**, prismalayanoside **54**, 3β-hydroxyolean-12-en-28-*O*-acetoate **44**, lucidin-3-methyl ether **55**, rubiadin-1-methyl ether **17**, damnacanthol **56**, rubiadin **12**, 1-ethyl-3-hydr oxymethyl-4-hydroxy-9,10-anthraquinone **57**, 2-methoxy-3-oxyhydroxymethyl-9,10-anthraquinone **58**, lucidin-ω-methyl ether **14**, and asperuloside**26**.[25]

P. tetrandra (Roxb) K. Schum

P. tetrandra is locally known as *tongkata jisamat* and widely distributed in South East Asia such as India, Sri Lanka, Bangladesh, Vietnam, Thailand, and Peninsular Malaysia.

The earliest report was by Tu *et al*. they have successfully isolatedtectoquinone **18**, rubiadin **12**, rubiadin-1-methyl ether **17**, damnacanthal **40**, β-sitosterol **42**, ursolic acid **48**, and daucosterol **62**.[20] Feng *et al*. continued their phytochemical studies on this species and successfully identified three new anthraquinones; 1-hydroxy-2,3-dimethoxy-7-methyl-9,10-anthraquinone **11**, 1,3-dihydroxy-5,6-dimethoxy-2-methyl-9,10-anthraquinone **38**, and 3-hydroxy-1,5,6-trimethoxy-2-methyl-9,10-anthraquinone **63**, together with five known compounds, namely 1-hydroxy-2-methyl-9,10-anthraquinone **47**, 1,3-dihydroxy-2-methoxy-9,10-anthraquinone **27**, 1,3-dihydroxy-2-methyl-9,10-anthraquinone **12**, 3-hydroxy-1-methoxy-2-methyl-9,10-anthraquinone **17**, and 2-hydroxy-3-hydroxymethyl-9,10-anthraquinone **64**, from the root extracts.[6] In addition, Krohn *et al*. managed to isolate a new complex iridoid, prismatomerin **65**, together with the known glucoside gaertneroside **66** from the leaves extracts.[14] Recently, Abdullah *et al*. had further investigated this species and obtained three pentacylic triterpenoids; namely ursolic acid **48**, 3β,19,23-trihydroxyurs-12-en-28-oic acid **67**, and 3β-acetylolean-12-en-28-oic acid **43**.[26]

P. glabra (Korth.) Valeton

P. glabra is a tropical plant, grows on hillsides and ridges of tropical forests at altitudes up to 700 m in Peninsular Malaysia, Sumatra, and Borneo.[19,27] Only one literature about this plant in 2013 when Mohamad *et al*. described the isolation of four anthraquinones, which are 1,3-dihydroxy-2-methoxymethyl-9,10-anthraquinone **14**, 1-hydroxy-3-methoxy-2-(methoxymethyl)-9,10-anthraquinone **59**, 2-methyl-3-methoxy-9,10-anthraquinone **60**, and 1,3-dimethoxy-2-methyl-9,10-anthraquione **61**.[11]

P. sessiliflora Pierre ex Pitard

P. sessiflora is a tree growing in Thailand, Laos, Cambodia, and Vietnam. This is the only report refers to the identification of rubiadin **12** and rubiadin-1-methyl ether **17** from the methanol extract of the roots.[3]

BIOLOGICAL ACTIVITIES

Cytotoxicity Activity

Hao *et al*. have reported the cytotoxicity of the anthraquinone glycosides **1-7** isolated from *P. connate* using human lung cancer (A549) and human hepatoma (HepG2) cells grown in RPMI-1640 medium plus 10% heat-inactivated fetal bovine serum by MTT method. The aglycone**7** of **4** exhibited a potent inhibitory effect on both A549 and HepG2 cell lines with IC_{50} of 6.72 and 9.38 μmol/L, respectively. However, all six anthraquinone glycosides **1-6** nearly inactive (IC_{50}>100 μmol/L) against the cell lines. Compound **9** exhibited potent cytotoxicity against the A549 and lung adenocarcinoma (LAC) human cancer cell lines with IC_{50} values of 4.5 and 7.8 μM, respectively.[13] Compounds **13-16**, **18**, **23**, and **24** showed inhibitory effects against A549 cell line with IC_{50} values ranging from 16.1 to 93.6 μM and compounds **11-17**, **18**, **23**, and **24** exhibited activities against LAC cell line with IC_{50} values ranging from 9.6 to 99.1 μM. The activity profiles suggested that the phenolic OH group might be necessary for the antitumor potency of tetrahydroanthraquinones and anthraquinones.[7] The cytotoxicity of compounds **31-37** was tested in a panel of human lung tumor cells (H1229, HTB179, A549, and H520 cell lines). Compound **37** showed the strongest activity in the suppression of tumor cell growth (IC_{50} 2.7–3.9 μM) and the others with chelated phenolic hydroxyls exhibited relatively lower activity (IC_{50} 8–20 μM). The activity profiles of these compounds may be due to the positions of hydroxyl groups at C-5 and C-8, which might be required for their antitumor potency, and hydroxylation at C-1 could significantly enhance its cytotoxic activity.[12] Compounds **39**, **40,** and **41** isolated from *P. fragrans* exhibited cytotoxic activity against breast cancer (BC) cell line with respective IC_{50} values of 6.9, 10.1, and 8.0 μg/mL. In addition, compounds **38**, **44**, **39**, **40**, **12**, **17**, and **43** showed cytotoxic activities against NCIH187 cell line with IC_{50} values of 8.7, 5.1, 1.9, 13.5, 14.2, 4.5, and 9.4 μg/mL, respectively.[15] Compounds **65** and **66** isolated from *P. tetrandra* were tested for cytotoxicity in the brine shrimp lethality assay. Both compounds showed remarkable toxicity (LD_{50} 72 ng/mL (195 μM) for **65** and 156 μg/mL (380 μM) for **66** at 24 h, and LD_{50} below detection limit (all shrimps dead) for **65** and 92 μg/mL (224 μM) for **66** at 48 h.[14]

Anticancer Activity

Compound **17** isolated from *P. connate* possesses good anticancer activity toward BC cell lines (IC_{50} of 4.05 μg/mL) and was inactive against KB cell lines (IC_{50} of 13.12 μg/mL), whereas compounds **17** and **40** were inactive for both cell lines (IC_{50} of >20 μg/mL) (9). Besides, compound **65** isolated from *P. tetrandra* was tested against NCI in an *in vitro* primary anticancer assay, a three-cell-line panel comprising MCF7 (BC), NCl-H460 (lung cancer), and SF-268 (central nervous system cancer). The cells showed zero growth in these tests in the presence of compound **65**.[14]

Antifungal Activity

The hexane and CH_2Cl_2 extracts of air-dried roots and stems of *P. fragrans* exhibited antifungal activity toward *Candida albicans* with IC_{50} values of 11–17 μg/mL. Compounds **39** and **40** showed antifungal activity towards *C. albicans* with IC_{50} values of 6.0 and 22.6 μg/mL, respectively.[15] Both compounds also showed moderate activity against *C. albicans* with IC_{50} values of 5.18 and 5.58 μg/mL, respectively, whereas compound **17** was inactive (IC_{50} of >50 μg/mL).[9]

Antimalarial Activity

The root methanol extract of *P. sessiflora* has shown significant antimalarial potential against T9/94 *Plasmodium falciparum*. In addition, compounds **12** and **17** which isolated from this species revealed weak activity with IC_{50} range 1300 µg/mL and 1560 µg/mL, respectively.[3] Besides, compounds **17**, **39**, and **40** isolated from *P. malayana* did not show any antimalarial activity toward *P. falciparum* (IC_{50} of >10 µg/mL).[9]

Antiplasmodial Activity

The hexane and CH_2Cl_2 extracts of air-dried roots and stems of *P. fragrans* exhibited antiplasmodial activity toward *P. falciparum* with IC_{50} values of 3.1–3.7 µg/mL. Compound **43** isolated from this species was found to be active in this assay with an IC_{50} value of 5.9 µg/mL.[15]

Antitumor Activity

Compound **25** isolated from *P. connata* was examined for antitumor activity on human laryngocarcinoma HEp-2 cells *in vitro* and revealed that it is dramatically inhibited HEp-2 cell growth, induced the cell cycle arrest at the G2/M phase and efficiently induced cell apoptosis.[8]

Antituberculosis Activity

Compounds **39, 40, 12, 41,** and **43** which isolated from *P. fragrans* showed antituberculosis activity toward *Mycobacterium tuberculosis* with minimum inhibitory concentration (MIC) values of 100, 25, 100, 50, and 50 µg/mL, respectively.[15]

Antimycobacterial Activity

The hexane and CH_2Cl_2 extracts of air-dried root and stem of *P. fragrans* exhibited antimycobacterial activity toward *M. tuberculosis* with MIC values of 25–100 µg/mL.[15]

CONCLUSION

In this review, we synopsizeon the secondary metabolites isolated from the genus *Prismatomeris* and their pharmacological properties. Most of the species produced anthraquinones, iridoids, and triterpenoids. Apart from that, further phytochemical studies are needed in the near future to provide a more detailed pattern of the natural constituents and the biologically active principles in extracts. As a conclusion, it is manifest that the genus *Prismatomeris* comprises therapeutically promising and valuable plants in the ethnomedical traditions. Meanwhile, few studies described its pharmacological properties, and this genus merits more intention in continuing research for new bioactive compounds.

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