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A review on herbal remedies of various Genus *amoora*

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ABSTRACT

The genus *Amoora* belongs to the Meliaceae family comprised about 25–30 species. The plants of this genus are economically important timber tree, usually distributed in the tropical and subtropical regions of Asia, mostly in China, India, Malaysia and Bangladesh. Species of this genus are important ornamental ecological plants, which provide useful timber for building purposes. Several *Amoora* species have been used as folk medicines in Southeast Asia for the treatment of many diseases such as diarrhea, inflammation, spleen and liver and cardiac diseases. As the characteristic constituents of the Meliaceae family for structurally diverse and biologically significant limonoids have attracted increasing research attentions. In recent years, more and more bioactive chemical constituents have been found to validate the folk medicinal usages from many *Amoora* species. However, upto now no complete review has been published. In this present study review, we summarizes systematically their phytochemistry and pharmacological activities of genus *Amoora* reported in available literature as retrieved from PubMed and Google Scholar with aim of providing useful findings for further studies and reasonable utilization.

Keywords: *Amoora,* Maliaceae, Tropical, vermicide, Cardiac diseases.

1.INTRODUCTION

Plants of *Amoora* genus have been extensively used as important timber tree, which also are cultivated as ornamental ecological plants. Some *Amoora* species have been used as traditional medicine to treat dysentery, laxative, skin, cardiac diseases in several countries for a long period. The stem barks of *A. tsangii*, known as "Tie luo" in China, are mainly used as a vermicide in folklore medicine. Among *Amoora* species, *A.tsangii, A.dasyclada, A.rohituka, A.cucullata* and *A.tetrapetala* have been reported in traditional medicine as important folk herb.

Up to now 140 compounds (except volatile constituents) have been reported from nine *Amoora* species (*A.tsangii, A.dasyclada, A.rohituka, A.cucullata*, *A.tetrapetala, A.yunnanensis, A.stellato-squamosa, A.ouangliensis and A.cucullata*) including eight sesquiterpenoids (1–8), twenty-six diterpenoids (9–34), forty-two triterpenoids (35–76), twenty-two limonoids (77–98), seven steroids (99–105), seven alkaloids (106–112), seven rocaglamide derivatives (113–119), four flavonoids (120–123), four glycosides (124–127), two coumarins (128–129), nine phenols (130–138) and two organic acids and esters (139–140). Terpenoids are the predominant secondary metabolite constituents in the genus, chiefly triterpenoids, which may belong to the dammarane, tirucallane, cycloartane, taraxerane, oleanane, lupane and hopane skeletal types.

In this present study review, we summarize systematically their phytochemistry and pharmacological activities of genus *Amoora* reported in available literature as retrieved from PubMed and Google Scholar with aim of providing useful findings for further studies and reasonable utilization.

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2. VARIOUS GENUS TYPES OF AMOORA

2.1. *AMOORA CUCULLATA*

Amoora cucullata (Meliaceae) is a mangrove plant, distributed in the coastal areas of Southeast Asia and the Indian Ocean. This plant has been used by local Thai people as a folk medicine for treatment of marrow, diarrhea and inflammation.

As part of their continuing search for bioactive constituents from Thai medicinal plants separate hexane and dichloromethane soluble extracts of the fruits of *A. cucullata* were found to exhibit significant cytotoxic activity when evaluated against a panel of human cell lines. Fractionation of these extracts led to the isolation of two new naturally occurring cyclopenta[*b*]benzofuran, 1-*O*-formylrocagloic acid**(1)** and 3-hydroxyrocagloic acid**(2)** along with five known compounds called rocaglaol**(3)**, rocagloic acid**(4)**, 3 hydroxymethyl rocaglate**(5)**, 1-*O* formyl methylrocaglate**(6)** and methylrocaglate**(7)** from the fruits of *A. cucullata*. Their structures were elucidated by spectroscopic methods. Compounds 1,2,3,6 and 7 exhibited potent cytotoxicity against KB, BC and NCI-H187 cell lines whereas 4 and 5 showed selective cytotoxicity against NCI-H187 cell line. [1]

Ahmed F et al., as part of their continued research program into the isolation of bioactive compounds for overcoming TRAIL resistance, they explored medicinal plants from Bangladesh. After initial screening of the extracts library, Methanol extract of leaves of *Amoora cucullata* (Meliaceae) was found to be potently active in overcoming TRAIL resistance. *Amoora cucullata* is a tall tree, grown in the coastal forests of Bengal, Burma, Malay Peninsula andamans and Borneo. It was collected from the Sundarbans Mangrove Forest, Bangladesh, where it is known as 'Amoor', 'Latmi' and 'Natmi' among the local people. Its leaves are traditionally used in the treatment of inflammation. Juice of the leaves is antibacterial and extensively used for the treatment of dysentery, skin diseases and in cardiac diseases. [2]

Crude Methanol extract of leaves were reported to show anti-inflammatory, antinociceptive, diuretic and CNS depressant activities. Previous chemical investigation of its leaves reported about isolated polyphenols and tannins, whereas from its fruits, several rocagloic acid derivatives were isolated. Bioassay-guided fractionation of Methanol extracts of its leaves led to the isolation of four new compounds along with seven known compounds. Of those isolates, compounds **1**, **5**, **8** and **9** showed TRAIL resistance-overcoming activity, among which **8** showed the most potent activity and enhanced TRAIL induced apoptosis in TRAIL-resistant human gastric adenocarcinoma (AGS) cells through the activation of caspase-3/7, enhancing the expression of DR4 and DR5 mRNA in AGS cells. Cell death caused by the combined treatment of **8** and TRAIL was inhibited by human recombinant DR5/Fc and DR4/Fc chimera proteins, indicating that **8** sensitizes TRAIL-resistant AGS cells to TRAIL through the induction of DR4 and DR5. [3]

The fresh leaves of *Amoora cucullata* were collected from the Sundarbans Mangrove Forests, Bangladesh. The methanolic extract of *A. cucullata* was partitioned successively with ethyl acetate, *n*-Butanol and water. The ethyl acetate fraction was then subjected to repeated column chromatography on Sephadex LH20, silica gel 60N, octadecylsilyl (ODS) gel and preparative thin layer chromatography (PTLC) to obtain dasyclamide (**2**), ent-2b –hydroxymanool (**3**), chrysin (**4**), apigenin (**5**) and kaempferol-3-*O*-b -D-glucopyranoside (**6**). All of the known compounds were identified by spectroscopic data including 1D and 2D NMR. Additionally, juice of the leaves found to possess antibacterial activity and extensively used for the treatment of dysentery, skin and cardiac diseases. The crude methanolic extract of leaves were reported to show anti-inflammatory, antinociceptive, diuretic and central nervous system (CNS) depressant activities. Phytochemical analysis of the stem bark of *Amoora cucullata* led to the isolation of fridelin, stigmasterol, b -sitosterol, betulinic acid and caffeic acid. Furthermore, analysis of the leaves showed the presence of several polyphenols and tannins, whether from its fruits, several rocagloic acid derivatives were isolated. Finally, in continuation of their phytochemical screening of *A. cucullata* leaves, they report the isolation and structure elucidation of one new putrescine bisamide, cucullamide (**1**), along with five known natural compounds (**2—6**). [4]

2.2. AMOORA DASYCLADA

The Meliaceae family comprises about 1400 species, mainly distributed in tropical and subtropical areas, with 62 among them being found in China. Plants of this family are rich sources of structurally diverse and biologically significant limonoids. *Amoora dasyclada*, a member of the Meliaceae family, yields varied kinds of terpenoids, including diterpenoid, triterpenoid and tetraterpenoid, with anticancer and cytotoxic activities. They firstly isolated three new inositol angelates and two alkaloids which have not been investigated previously in *Amoora dasyclada*. Angelate compounds were exhibited to have antitumor activity: 3-ingenyl angelate has significant inhibitory effects on skin cancer; decursinol angelate caused a significant increase in the life span and a significant decrease in the

tumor weight and volume of mice inoculated with Sarcoma-180 tumor cells. Tirucallane derivatives with a pyrrole ring from Meliaceae family showed obvious cytotoxicity against five human cancer cell lines. Compounds **1–5** were identified by 1D and 2D NMR method and HR-ESI-MS and structurally compared with similar compounds found previously. [5]

Three new inositol angelate compounds (**1–3**) and two new tirucallane-type alkaloids (**4** and **5**) were isolated from the *Amoora dasyclada* and their structures were established. Compounds **4** and **5** exhibited significant cytotoxic activity against human cancer cell line HepG2 with IC50 values at 8.4 and 13.2 _M. In addition, compounds **4** and **5** also showed remarkable toxicity to the larvae of Artemia salina. [5]

Two novel tirucallane triterpenoids (**4** and **5**) with cytotoxicity identified in this research can provide evidence of chemotaxonomy and medicinal value for this species. Additionally, the rarely studied inositol angelates had only been isolated from the Chinese traditional medicine herb Inula cappa before, which is an ingredient of a famous Chinese formula against fever, abdominal distention and menoxenia. Identification and evaluation of three new inositol angelates (**1–3**) from *Amoora dasyclada* in this work suggest the further definition of the roles of *Amoora dasyclada* in phytochemistry research and medicinal application. [5]

A new tetranortriterpene 3α-acetoxy-24, 25, 26, 27-tetranortirucalla-7-ene-23(21)-lactone (**3**) and eleven other compounds were isolated from the twigs of *Amoora dasyclada*. The structure of compound **3** was identified on the basis of spectroscopic data and the bioactive experiments of **1** and **3** D**5** against AGZY 83-a (human lung cancer cells) and SMMC-7721 (human liver cancer cells) are documented. Among them, compound **5** exhibited a strong activity against SMMC-7721(human liver cancer cells). [6]

2.3 AMOORA OUANGLIENSIS AND STELLATO SQUAMOSA

The barks of *A. ouangliensis* and the twigs of *A. stellato-squamosa* were collected in Xishuangbanna County of Yunnan Province, P. R. China, in January 2002. The plants were identifi ed by Prof. Jing-Yun Cui, Xishuangbanna Tropical Botanical Garden, Academia Sinica, Mengla County, China. This study presents a new *ent*-halimane-type diter pene, 5(10), 14- *ent*-halimadien-3β, 13S-diol (**1**), obtained during our continuing study on the bark of *Amoora ouangliensis*. Ten other diterpenoids, namely neoclerod- 14-en-3α,4β,13*S*-triol (**2**), 6-*O*-acetyl-austroinulin (**3**), (3α,4β)-neoclerod-13(*E*)-en-3,4,15-triol (**4**), 3,13(*E*)-2-oxo-neoclerodadien-15-ol (**5**), methyl (13*E*)-2-oxoneocleroda-3**(6)**,13-dien-15-oate**(7)**, (13*S*)-2-oxoneocleroda-3,14-dien-13-ol (**8**), (13*E*)-neocleroda-3,13-dien-15,18-diol (**9**), 15-hydroxy-8(17),13(*E*)-labdadien-19-oic acid (**10**), 8(17),12(*E*),14-labdatrien-19-oic acid (**11**), (3α,4β, 14*RS*)-neoclerod-13(16)-en-3,4,14,15-tetrol (**12**), were isolated from the barks of *A. ouangliensis* and the twigs of *A. stellato-squamosa*. The bioactive experiments on **1** – **13** (**6** and **13** were the acetylized products of **4** and **12**, respectively against AGZY 83-a (human lung cancer cells) and SMMC-7721 (human liver cancer cells) were also assayed. Among them, **1** and **6** exhibited significant activities against these cells with IC50 values of 21.52 and 28.47 μM and 25.73 and 23.34 μM respectively. [7]

2.4. AMOORA TSANGII

Limonoids are a large class of nortriterpenoids that are mainly found in plants belonging to the Meliaceae family. To date, more than a thousand limonoids have been reported. They can be divided into four major groups, i.e., ring intact limonoids, seco-limonoids, rearranged limonoids and limonoid derivatives. Nitrogen-bearing limonoids, which are also known as limonoid-based alkaloids, are a minor class of limonoids. These compounds consist of five lactam-bearing, four pyridine-bearing and two maleimide-bearing limonoids. [8]

Amoora tsangii is a meliaceous timber tree native to Hainan Island in China. The stem bark of this plant is used as a folk medicine to kill lice. A previous phytochemical investigation of the twigs and leaves of *A. tsangii* reported nine limonoids and four sesquiterpenoids. Their preliminary LC-MS study of the ethanolic extract of *A. tsangii* revealed the presence of several N-bearing limonoids, which prompted us to conduct an extensive study of limonoids in this plant. They describe the isolation and structural elucidation of 12 new bearing limonoids and the inhibitory activity of a selection of these compounds toward NF-κB. [8]

The twigs and leaves of *A. tsangii* were extracted twice with 80% EtOH and the combined extracts were concentrated under reduced pressure, suspended in H_2O and extracted sequentially with petroleum ether and EtOAc. The EtOAc-soluble fraction was repeatedly purified by column chromatography over silica gel and ODS and by preparative HPLC to afford the N-bearing limonoids 1–12.

Twelve new lactam-bearing limonoids, amooramides A–L (1–12) were isolated from the twigs and leaves of Amoora tsangii and their structures were fully elucidated by spectroscopic analysis. These compounds represent the first example of rings A,B-seco-limonoids bearing

unusual lactam side chains at C-17, including a 3-substituted 1,5-dihydro-2Hpyrrol-2-one moiety in 1–7 and a 4-substituted 1,5-dihydro-2H-pyrrol-2-one unit in 8–12. Compound 9 inhibited TNFα-induced NF-κB activity by 64% at a concentration of 10 μM. [8]

The twigs and leaves of *Amoora tsangii* were collected from Hainan Island of China in February 2004 and authenticated by Prof. Shi-Man Huang, Department of Biology, Hannan University of China. The air-dried powder of twigs and leaves of *A. tsangii* (9 kg) was extracted three times with 95% Ethanol at room temperature to provide an extract (800 g), which was partitioned between Ethyl acetate and water to give an Ethyl acetate soluble fraction (165 g). Their structures were elucidated primarily on the basis of spectroscopic data. Seven new limonoids, amotsangins A-G (**1**-**7**), two known limonoids and four known sesquiterpenoids were isolated from the twigs and leaves of *Amoora tsangii*. [9]

2.5. AMOORA ROHITUKA

Amoora rohituka is described in Ayurveda, an Indian traditional system of medicine for management of disorders of blood, diseases of eye, helminthiasis disease, ulcer, liver disorders and splenomegaly. However, the leaves were not reported to have anticancer properties till date. This study was carried out to evaluate the cytotoxic potential of leaf extracts of Amoora rohituka. The leaves powder was macerated in petroleum ether, ethyl acetate and methanol and evaluated their anticancer activities in vitro. The phytochemical constituents of ethyl acetate extract were screened by FTIR analysis and phytochemical screening methods. [10]

The ethyl acetate extract (RLEA) showed the presence of alkaloids, flavonoids, steroids, tannins, saponins and terpenoids. The RLEA exhibited high cytotoxic effect against human breast cancer cells, MCF-7 (IC50 ¼ 9.81 mg/mL) and induced apoptosis by altering nuclear morphology and DNA laddering. Wound healing assays explained the potency of extract to decrease the cell migration. The extract of Amoora rohituka leaves exhibited anticancer activity with less toxicity and it could be used for development of alternative drugs in the treatment of human breast cancer. [10]

Earlier studies on this plant had disclosed the presence of aphanamixinin and sterol, saponins, flavanone and anthraquinone glycosides. Further study of this plant has now led to the isolation of one congener which biogenetically links a class of limonoids andirobin. A new limonoid named amoorinin dihydroandirobin was reported on the basis of spectral and chemical methods. [11]

The plant *Amoora rohituka,* of the Meliaceae family, is a large evergreen tree of India with a straight cylindrical trunk 50 ft long and 5-6 inch in girth and is distributed over the sub-Himalayan tract of that country. According to earlier reports, the seeds of this plant (78% kernel) yield ca. 47% of a reddish brown oil constituted of 57.4% linoleic and 11.2% oleic acids. One of the seed oils of the Meliaceae family, *Melia azadirachta,* which has already gained considerable commercial importance in this country, is composed of 49.1-61.9% oleic acid and 9.0-15.8% linoleic acid. Such wide variations in the compositions of the seed oils from two species of the same family warranted further investigation of the former by modern techniques. Also, the seed oil of *A. rohituka* appeared to be of interest to us in our study of the triglyceride composition of linoleic rich oils. [12]

On extraction with petroleum ether (bp 60-80 C), seed kernels of *A. rohituka* yielded 35% of deep yellow colored oil (18.4% free fatty acid) which on refining became light yellow in color. The fatty acid composition of the 2-monoglyceride obtained from lipolysis of the *A. rohituka* seed oil was determined by GLC analysis of its methyl esters. [12]

The triglyceride composition of *A. rohituka* seed oil calculated from the fatty acid composition of the original triglyceride and the 2-monoglyceride using the assumptions of Vanderwal et al. and Coleman et al., was found to be GS 3 2.2, GS2U 28.6, GSU 2 48.1 and GU 3 21.1 percents mole, respectively. [12]

Mutations of the K-Ras gene occur in over 90 % of pancreatic carcinomas and to date, no targeted therapies exist for this genetically defined subset of cancers. STAT3 plays a critical role in KRAS-driven pancreatic tumorigenesis, suggesting its potential as a therapeutic target in this cancer. Therefore, finding novel and potential drugs to inhibit oncogenic K-Ras is a major challenge in cancer therapy. In an attempt to develop novel anti- KRAS mutant chemotherapeutics, they isolated three novel triterpenoids from Amoora rohituka stem and their chemical structures were characterized by extensive 1H-NMR, 13CNMR, Mass, IR spectroscopic studies and chemical transformations. Aphanin (3alpha-angeloyl oxyolean-12-en-28-oicacid) is one of the isolated novel triterpenoid compounds. They found aphanin exhibited anti-proliferative effects, caused G0-G1 cell cycle arrest, inhibits K-RasG12Dmutant activity by decreased STAT3, p-STAT3, Akt, p-Akt, cyclinD1and c-Mycexpressions and induced apoptosis in pancreatic cancer HPAF-II (ΔKRASG12D) cells. The apoptosis proceeded

through depletion of GSH with a concomitant increase in the reactive oxygen species production. The results of the study shown important implications for the development of aphanin as potential novel agent for the treatment of K-Ras mutant pancreatic cancer and STAT3-cMyc-cyclinD1 axis may serve as an important predictive biomarker for the therapeutic efficacy. [13]

tRohitukine, a chromone alkaloid, has gained considerable international attention in recent years because of its novel semi-synthetic derivative, flavopiridol and P-276-00. Both these molecules are in advanced stages of clinical development and trial for cancer treatment. Recently, flavopiridol was approved as an orphan drug for treatment of chronic lymphocytic leukemia cancer. The natural occurrence of rohitukineis restricted to only four plant species, Amoora rohituka and Dysoxylum binectariferum and from Schumanniophyton magnificum and Schumanniophyton problematicum. Recently, an endophytic fungi isolated from D. binectariferum was reported to produce rohitukine in culture. In this study, it has been reported that the production of rohitukine and its subsequentattenuation by endophytic fungi, Fusarium oxysporum (MTCC-11383), Fusarium oxysporum (MTCC-11384) and Fusarium solani (MTCC-11385), all isolated from D. binectariferum and Gibberella fujikuroi (MTCC-11382) isolated from Amoora rohituka. The fungal rohitukine which was analyzed by HPLC, LC–MS and LC–MS/MS was identical to reference rohitukine and that produced by the plant. The rohitukine content in the mycelial samples ranged from 192.78 g to 359.55 g 100 $g^{-1}$of dry weight and in broth it ranged from 14.10 to 71.90 g 100 ml^{-1}. In all the fungal cultures, the production declined from first to fourth sub-culture. Studies are underway to unravel the mechanism by which the fungi produce the host metabolite in culture. [14]

Toona ciliate and *Amoora rohituka* stem bark were collected from Comilla in August 2000. The plants were identified at the Bangladesh National Herbarium where voucher specimens have been deposited under the accession numbers DACB-28926 and DACB-28927, respectively. *A. rohituka* is feeding deterrent activity against *Tribolium castaneum* (Coleoptera: Tenebrionidae) of some secondary metabolites w6x, hepatoprotective activity of the plant extract w7x, antiviral and antibacterial activity of the isolated limonoid rohitukin w8x, cytotoxicity of amoorastatin w9x, growth inhibitory effect of 12a-hydroxyamoorastatin against murine P388 lymphocytic leukaemia cell lines w9x. [15]

The petroleum ether and dichloromethane extracts of *A. rohituka* yielded zones of inhibition of 07–19.5mm at 500 mg disc while the methanol extract demonstrated prominent antibacterial activity with zones of inhibition of 11–30 mm at the same dose. From the test results of antifungal screening it appears that the extracts have fungitoxicity against all the test pathogens in a different degree but not in a significant extent. [15]

Chemotherapeutic agents for cancer are highly toxic to health tissues and hence alternative medicine avenues are widely researched. Majority of the recent studies on alternative medicine suggested that *Amoora rohituka* possesses considerable antitumor and antibacterial properties. In this work, *rohituka* and *chittagonga*, fractionated with petroleum ether, dichloromethane and ethanol, were explored for their anticancer potential against two breast cancer (MCF-7 and HTB-126) and three pancreatic cancers (Panc-1, Mia-Paca2 and Capan1). The human foreskin fibroblast, Hs68, were also included. Cytotoxicity of each extract was analyzed using the MTT assay and label-free photonic crystal biosensor assay. A concentration series of each extract was performed on the six cell lines. For MCF-7 cancer cells, the *chittagonga* (Pet-Ether and CH2Cl2) and *rohituka* (Pet-Ether) extracts induced cytotoxicity; the *chittagonga* (EtoAC) and *rohituka* (Methanol) extracts did not induce cytotoxicity. For HTB126, Panc-1, Mia-Paca2 and Capan-1 cancer cells, only the *chittagonga* CH2Cl2 extract showed a significant cytotoxic effect. The extracts were not cytotoxic to normal fibroblast Hs68 cells, which may be correlated to the specificity of *Amoora* extracts in targeting cancerous cells. Based on these results, further examination of the potential anticancer properties *Amoora* species and the identification of the active ingredients of these extracts are warranted. [16]

3. CONCLUSION

The genus of *Amoora* in its literature survey shows various claims of therapeutic activity for a variety of ailments. However, these claims are unauthenticated by the scientific proof. They also possessed chemical constituents with diverse structural types, exhibited extensive pharmacological activities. Therefore, *Amoora* species seems to have great potential as a promising ethnopharmacological plant source. However, majority of its species chemical constituents remain unknown, which restrict their utilization and development. The crude extracts of

Amoora species and their chemical constituents were found to possess extensive pharmacological activities, many chemical constituents have never been pharmacologically tested. Comprehensive investigations on the genus *Amoora* should be fully strengthened to clarify their chemical constituents and pharmacological effects as well as their relationships between the species in the near future. This review may provide useful information for further proper utilization of *Amoora* species as folklore medicines and future drug discovery in research field.

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