Biological activities and chemical constituents of some mangrove species from Sundarban estuary: An overview

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ABSTRACT

This review represents the studies performed on some beneficial mangrove plants such as *Ceriops decandra, Xylocarpus granatum, Xylocarpus moluccensis, Excoecaria agallocha, Sarcolobus globosus, Sonneratia caseolaris* and *Acanthus ilicifolius* from the Sundarban estuary spanning India and Bangladesh with regard to their biological activities and chemical investigations till date. Sundarban is the largest single chunk of mangrove forest in the world. The forest is a source of livelihood to numerous people of the region. Several of its plant species have very large applications in the traditional folk medicine; various parts of these plants are used by the local people as cure for various ailments. Despite such enormous potential, remarkably few reports are available on these species regarding their biological activities and the active principles responsible for such activities. Though some chemical studies have been made on the mangrove plants of this estuary, reports pertaining to their activity-structure relationship are few in number. An attempt has been made in this review to increase the awareness for the medicinal significance as well as conservation and utilization of these mangrove species as natural rich sources of novel bioactive agents.

Key words: Activity-structure relationship, bioactivities, folk-medicine, mangroves, phytochemicals, Sundarban estuary

INTRODUCTION

Mangroves are woody, specialized types of trees growing in brackish wetlands in the tropical and sub-tropical inter-tidal coastal zones and river deltas where other plants cannot grow. There are about 39.3 million acres of mangrove forests in the warm coastlines of tropical oceans all over the world. Among these the Sundarban (latitude 21° 31' - 22° 30' North and longitude 88° 10' - 89° 51' East) is the largest single block of tidal halophytic mangrove forest in the world.[1] The name Sundarban, literally meaning "beautiful forest" is believed to be derived from Sundari or Sundri (*Heritiera fomes*), one of the most abundant tree species found in this forest.[2] The forest lies in the Ganges-Brahmaputra Delta along the Bay of Bengal and is spread across areas of Bangladesh and West Bengal, India, forming the seaward fringe of the delta. The seasonally flooded Sundarban freshwater swamp forests lie inland from the mangrove forests. Of the total 10,000 km^2 area, Indian Sundarban mangrove forest covers 4,266.66 km^2 and the rest is covered with Bangladeshi Sundarban mangrove forest.[2] The biodiversity of Sundarban mangrove forest is rich and wide-ranging for which both of its Indian and Bangladeshi parts have been declared as World Heritage Site by UNESCO in 1987 and in 1997 respectively.[2,3] Mangroves thrive in extremely stressful and hostile environment of high salinity, high and low tides of water, high temperature and moisture, strong winds and muddy anaerobic soil. Besides these abiotic stress conditions, factors such as insects and microorganisms contribute largely in developing the biotic stress to the community. There is probably no other group of plants with such highly developed morphological and physiological adaptations to such extreme conditions. To thrive in such hostile environments, alterations in their physiological processes have occurred resulting in the synthesis of novel chemical compounds; these chemical compounds offer protection to these plants against various biotic and abiotic stresses mentioned above.[4-6] A number of these compounds or secondary metabolite molecules have significant biological and other medicinal properties that can be exploited in shaping better human health care needs. In fact, many mangrove plant species have their uses in folk

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or traditional medicine as cures for various ailments and for other commercial purposes.[7] These chemical compounds derived from the natural sources such as the mangroves can play very significant roles in the new drug discovery process.[8] However, intense scientific works need to be carried out to delve deeper into this scantily explored yet promising area to unfold the rich sources of valuable elixir for the health care of mankind.

The present review attempts discussion on the biological activities and chemical investigations carried out on mangrove plants *Ceriops decandra*, *Xylocarpus granatum, Xylocarpus moluccensis, Excoecaria agallocha, Sarcolobus globosus, Sonneratia caseolaris* and *Acanthus ilicifolius* from the Sundarban estuary spanning India and Bangladesh.

Ceriops decandra

C. decandra (Griff.) Ding Hou (Rhizophoraceae), locally called Jale Goran or Jhamti Goran in Bengali, is an evergreen shrub or small tree upto 5 m tall and are common in Indian Sundarbans.[2] The whole plant is used as an astringent and the plant parts are used to stop hemorrhage and treat ulcers, pain and hepatitis.[7,9] Though a few chemical investigations on *C. decandra* from other parts of India and the globe revealed the presence of diterpenoids, triterpenoids and lignins,[10-14] remarkably little studies on the chemistry and the biological activity of the species from Indian or Bangladeshi Sundarbans have been reported till date. Study by Ghosh *et al*.,[15] revealed significant contents of lipids, sterols and triterpenes from the leaves of *C. decandra* from Indian Sundarban region. The study also revealed the sterol and triterpene composition of the leaves of the species. Misra *et al*.,[16] reported the hydrocarbon and wax ester profile from the leaves of this species from Indian Sundarban along with six other species of mangroves. Uddin *et al*.,[9] reported the antinociceptive activity of ethanol extract of leaf and pneumatophore of the species from Bangladeshi Sundarban exhibiting significant inhibition of acetic acid-induced writhing in mice. A study on the leaf, bark and pneumatophore from the species of Bangladeshi origin showed potent broad spectrum activity against both Gram-positive and Gram-negative bacteria.[17] Another study by Banerjee *et al*.,[18] demonstrated strong antioxidant activity of the stem bark extract (reducing power as Ascorbic acid equivalent = 13.04 mg/g and diphenyl picryl hydrazyl [DPPH] radical scavenging ability as IC_{50} = 0.65 mg/ml) of *C. decandra* (Perr.) Robinson from Indian Sundarban. A recent study by Hossain *et al*.,[19] showed the antioxidative property of bark of the species from Bangladeshi Sundarbans which is in line with a previous study by Banerjee *et al*.[18] The study also demonstrated significant anti-inflammatory activity by the species. Chaudhuri and Guha[20] reported the presence of antifungal activity of the leaf and fruit extracts of the species against *Fusarium oxysporum*. Another recent study by Simlai and Roy[21] reported the phytochemical contents and the antimicrobial activity of *C. decandra* extracts and the stability of this activity against extreme thermal and pH treatment. The study also reported the partial identification of the nature of the active constituents exhibiting the antimicrobial activity using thin layer chromatography (TLC)-fingerprinting technique.

Xylocarpus granatum

X. granatum Koeing (Meliaceae), locally called Dhundul in Bengali, is an evergreen tree with gray bark and grows in the inter-tidal ridge forest and river bank.[2] Extracts of different parts of the plant are reported to be used traditionally as relief for fever including one caused by malaria, inflammation, dysentery, diarrhea, cholera and other abdominal problems in certain parts of the globe.[3,7] Due to its quality reddish black colored timber, the wood is exploited for carpentry works, resulting in the scarcity of the species in the Indian part of the Sundarban.[2] The bark of the plant is used for tanning and for the preparation of amber dyes as well.[22] Investigation on the extracts of Bangladeshi *X. granatum* revealed potential for central nervous system (CNS) depressant activity and DPPH radical scavenging activity.[23,24] Chemical investigation by Wangensteen *et al*.[25] from the bark of *X. granatum* of Bangladeshi Sundarban revealed the presence of four previously reported limonoids [Figure 1], i.e., xyloccensin O (1), xyloccensin *P* (2), xyloccensin Q (3) and gedunin (4). The study also revealed the presence of two flavonoids, catechin (5) and epicatechin (6) and procyanidins of the B1 (7), trimer (8) and pentamer (9) type with catechin as the starter and epicatechin as the extender. Evaluation of the antioxidant activity (DPPH radical scavenging and 15-lipoxygenase [15-LO]) demonstrated high activity in case of catechin and procyanidins, of which procyanidin of the pentamer type was found to possess the maximum activity (IC_{50}: DPPH: 3.3 ± 0.3 μM, 15-LO: 9 ± 1 μM) [Table 1]. This is in line with another study carried out by Wangensteen *et al*.[3] Uddin *et al*.[26] isolated and identified two limonoids [Figure 1], gedunin (4) and 1α-hydroxy-1,2-dihydrogedunin (10) from the bark of *X. granatum* of Bangladeshi Sundarban and reported the anticolon cancer activity of gedunin (4) [Table 1]. The IC_{50} value for cytotoxic potential of gedunin (4) against CaCo-2 colon cancer cell line was found to be 16.83 μM. This exhibits the significant anticancerous property of the compound. Antidiarrheal property of its bark of Bangladeshi Sundarban origin has also been reported thus justifying its use in the traditional herbal medicine.[22] The methanol extract of the bark at oral doses of 250 mg/kg and 500 mg/kg showed significant antidiarrheal activity in the castor oil and magnesium sulphate induced murine models respectively. Daula and Basher[27] demonstrated the plant rootlet and shoot growth inhibitory activity as well as antimicrobial activity of the species from Bangladeshi Sundarban. The antimicrobial potential of the plant is in line with the studies reported by Alam *et al*.,[28] and Wangensteen *et al*.[3] Potent broad spectrum antibacterial activity against both Gram-positive and Gram-negative bacteria was also reported by Uddin *et al*.[17] from this species of Bangladeshi origin.

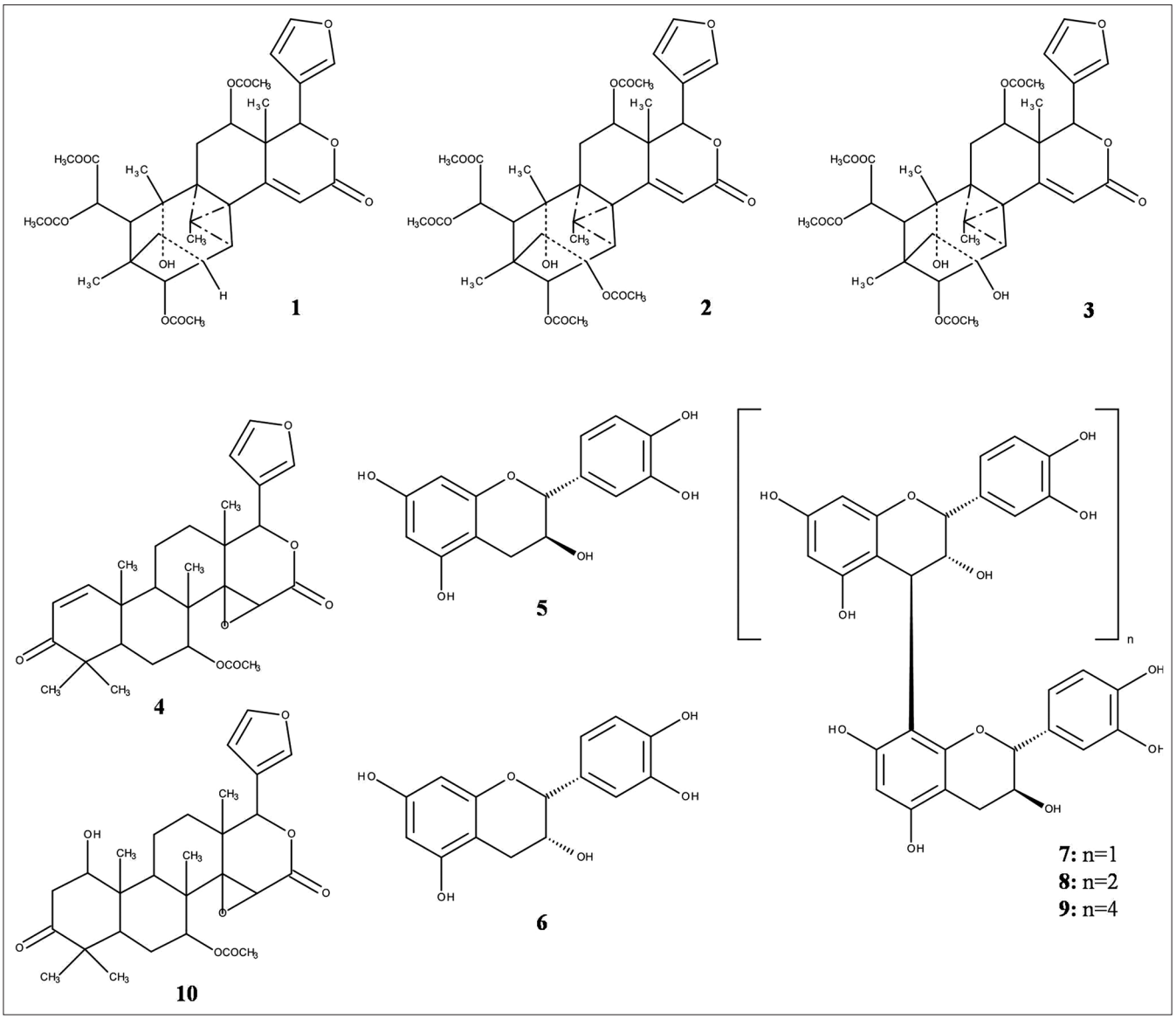

Figure 1: Structures of compounds isolated from *Xylocarpus granatum* (1-10)

Table 1: Biological activities of the compounds reported to be isolated from the mangrove plants of Sundarban estuary

Plant species	Isolated compounds reported to possess biological activity	Biological activities of the isolated compounds
Xylocarpus granatum	Xyloccensin O (1), xyloccensin *P* (2), gedunin (4), catechin (5), epicatechin (6), procyanidin B1 (7), procyanidin trimer (8), procyanidin pentamer (9)	DPPH radical scavenging activity (compounds 1, 2, 4-9);[3,25] 15-LO activity (compounds 1, 2, 5-9);[3,25] anti-colon cancer activity (compound 4)[26]
Xylocarpus moluccensis	Procyanidin trimer (15), procyanidin pentamer (16), procyanidin hexamer (17), procyanidin decamer (18), procyanidin undecamer (19)	DPPH radical scavenging activity (compounds 15-19);[3] 15-LO activity (compounds 15, 17-19)[3]
Sarcolobus globosus	Sarcolobin (20), tephrosin (22), 12aα-hydroxydeguelin (24), 11-hydroxytephrosin (25),12a-hydroxyrotenone (26), 6aα,12aα-12a-hydroxyelliptone (27), 6a, 12a-dehydrodeguelin (28), barbigerone (30), 6,7-dimethoxy-2,3-dihydrochromone (31), genistin (34), vanillic acid 4-O-β-D-glucoside (35), glucosyringic acid (36), tachioside (37), isotachioside (38)	DPPH radical scavenging activity (compounds 20, 22, 25-27, 31, 34, 37, 38);[3,25] 15-LO activity (compounds 20, 22, 24-28, 30, 31, 34-38);[3,25] brine shrimp lethality (compounds 20, 22, 26)[3,25]
Sonneratia caseolaris	Luteolin (39), luteolin 7-O-β-glucoside (40)	DPPH radical scavenging activity (compounds 39, 40)[55]

DPPH=Diphenyl picryl hydrazyl, 15-LO=15-lipoxygenase

Xylocarpus moluccensis

X. moluccensis (Lamk.) Roem. (Meliaceae), a medium sized tree, generally grow away from the frequent tidal inundation.[2] Traditionally the bark of the plant is used to treat gastrointestinal disorders such as dysentery, diarrhea, fever including that from malaria and possesses astringent properties; the fruit is used as an aphrodisiac and used as a cure for elephantiasis and swelling of the breast.[3,7,29] Uddin *et al*.[30] showed the antidiarrheal activity of the methanolic extract of the bark of the plant from Bangladeshi Sundarban in the castor oil and magnesium sulphate induced mice and antibacterial property thus validating the plant's use in gastrointestinal disorders in traditional medicine. Alamgir *et al*.[23] and Sarker *et al*.[31] demonstrated the neuropharmacological property of *X. moluccensis* from Bangladeshi Sundarban exhibiting the CNS depressant activity in mice. Though no antimicrobial activity from leaf was observed by Uddin *et al*.[17], but Haque *et al*.[32] reported the presence of strong antimicrobial activities of crude extracts from stem bark and three isolated pure compounds (structures not elucidated) called XM-1, XM-2 and XM-3. The study also suggested the presence of cytotoxic activity towards brine shrimp nauplii, exhibited by the crude extract of Bangladeshi *X. moluccensis*. Mondal *et al*.[33] reported antimicrobial activity of pneumatophores of the species and suggested the species as an important source for antimicrobial compounds. The pneumatophore extracts of the plant of Bangladeshi (Sundarban) origin demonstrated moderate cytotoxic activity against human breast ductal carcinoma cells (MDA-MB-453S) and human gastric adenocarcinoma cells (AGS cell line).[29] In a recent study, a number of procyanidins have been reported from this plant by Wangensteen *et al*.[3] while chemically investigating the bark of the plant from Bangladeshi Sundarban. The study revealed the presence of flavonoids [Figure 2] catechin (11) and epicatechin (12) and few procyanidins, i.e. procyanidin B1 (13), procyanidin B3 (14), procyanidin trimer (15), procyanidin pentamer (16), procyanidin hexamer (17), procyanidin decamer (18) and procyanidin undecamer (19). Their studies revealed the presence of DPPH radical scavenging activity and 15-LO inhibiting activities in these isolated procyanidins and antimicrobial activities from the tissue extracts of this plant [Table 1].

Excoecaria agallocha

E. agallocha L. (Euphorbiaceae), locally called Geoa or Gneoa in Bengali, is a medium dioecious tree upto 15-20 m tall.[2] Parts of the plant possess medicinal properties and are used to treat epilepsy, conjunctivitis, dermatitis, hematuria, leprosy and toothache.[7] The species from Bangladeshi Sundarban is reported to possess neuropharmacological activity when tested on mice at higher dosage.[23] Subhan *et al*.[34] reported potent antinociceptive and gastroprotective effect of the crude ethanolic extracts of bark from *E. agallocha* of Bangladeshi Sundarbans. Another study by Subhan *et al*.[35] demonstrated a few biological activities such as neuropharmacological, antimicrobial and cytotoxicity effect of the ethanolic extract from the plant bark of Bangladeshi origin. In this study, the extract was found to possess potential effect on the CNS, exhibited significant antimicrobial activity and considerable

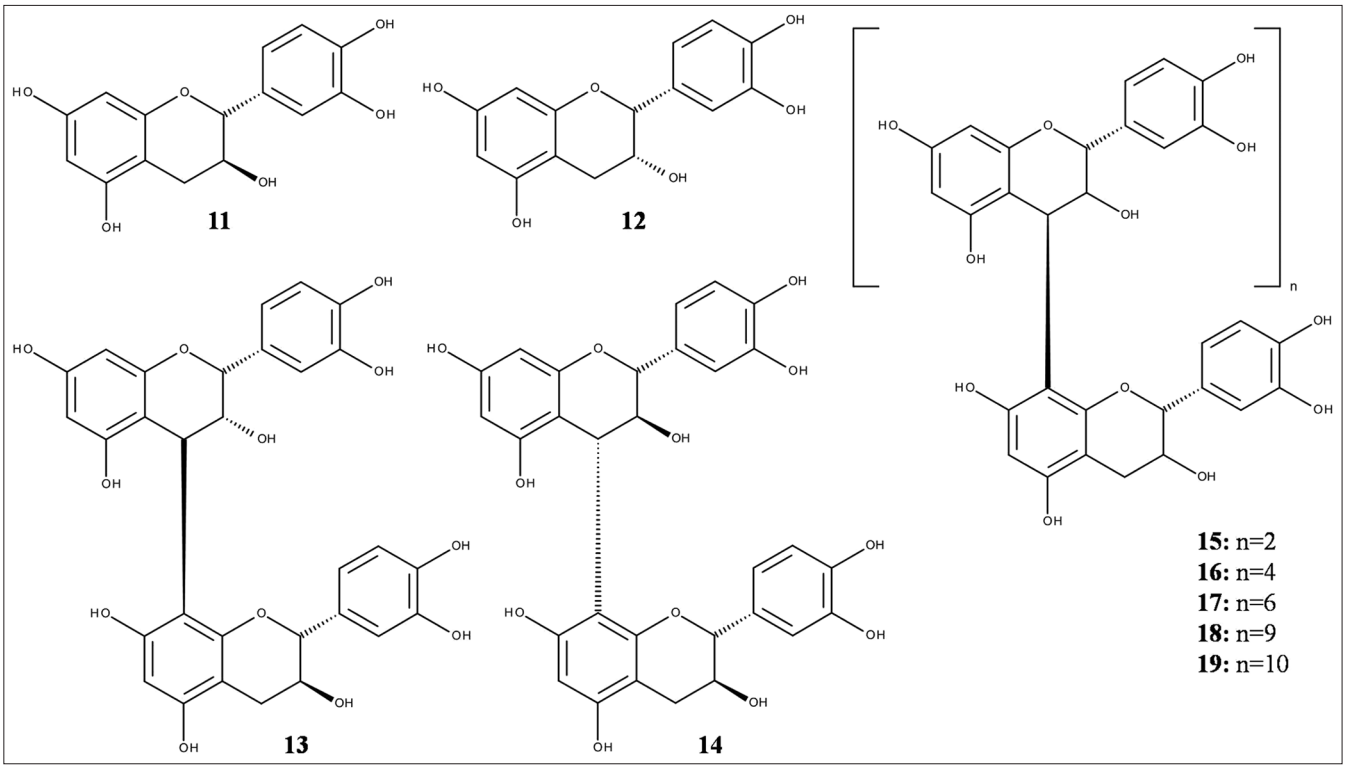

Figure 2: Structures of compounds isolated from *Xylocarpus moluccensis* (11-19)

cytotoxic effect on brine shrimps; however, the extract showed low level of toxicity in mice. The study by Subhan *et al*.[36] also demonstrated the presence of antioxidant activities in the tissue extracts of this plant from Bangaladeshi Sundarban. In another study, Hossain *et al*.[37] also reported the antioxidative property, along with the antiallergic property of different solvent extracts from the bark of the species of Bangladeshi origin. It was observed that the water and the ethanol fraction exhibited the maximum antioxidant and histamine release inhibitory activity compared to the other fractions. Kumar *et al*.[38] performed preliminary gas chromatography – mass spectrometry (GC-MS) analysis of the compounds present in the root exudates of the species from Indian Sundarbans. It is the first report on the presence of aminopyrine and palmitic acid in the root exudates of this species. A recent study by Rahman *et al*.[39] on the methanolic extract of stem from the species of Bangladeshi origin demonstrated more potent antihyperglycemic activity when compared to a standard antihyperglycemic drug, glibenclamide thus indicating the species as a potential source for antidiabetic drugs. Another study by Chaudhuri and Guha[20] showed that the water extract of bark of this species had antifungal activity against a pathogenic fungus, *F. oxysporum*. Despite these potential bioactivities, relatively little studies on the chemical content or compounds responsible for these activities of the species of Sundarban origin have been carried out. Some studies on the chemistry revealing the presence of phorbol ester, flavanone glycoside, various di- and triterpenoids in the *E. agallocha* from other parts of India and abroad have already been reported.[40-50] A very recent study by Mun *et al*.[51] revealed the chemical characteristics of *E. agallocha* along with few other mangrove species from Bangladeshi Sundarban demonstrating the dichloromethane, lignin, pentosan, α-cellulose etc., content in them.

Sarcolobus globosus

S. globosus Wall. (Asclepiadaceae), known as Caw Phal in Bengali, is a prostrate or climbing shrub growing in the mangrove forest of Sundarban estuary.[2] Traditionally, the plant is used as a relief for rheumatism, dengue and fever.[52] In a study carried out on the species from Bangladeshi Sundarban, Wangensteen *et al*.[52] for the first time reported the presence of a new rotenoid [Figure 3] sarcolobin (20) and a new isoflavone sarcolobone (21). The study also reported a few known rotenoids [Figure 3] such as tephrosin (22), 12aα-hydroxyrotenone (23), 12aα-hydroxydeguelin (24), 11-hydroxytephrosin (25), 12a-hydroxyrotenone (26), 6aα, 12aα-12a-hydroxyelliptone (27), 6a, 12a-dehydrodeguelin (28), 13-homo-13-oxa-6a, 12a-dehydrodeguelin (29), the isoflavone barbigerone (30) and a chromone 6,7-dimethoxy-2,3-dihydrochromone (31). The group for the first time reported 6,7-dimethoxy-2,3-dihydrochromone (31) as a natural product. Later on, Wangensteen *et al*.[53] identified two rotenoids, villosinol (32) and 6-oxo-6a, 12a-dehydrodeguelin (33), one isoflavone called genistin (34) and four phenolic glycosides named vanillic acid 4-O-β-D-glucoside (35), glucosyringic acid (36), tachioside (37) and isotachioside (38) for the first time from the species [Figure 3]. In this study, the rotenoids were found to inhibit 15-LO but lacked DPPH scavenging activity thus suggesting that the observed 15-LO inhibitory effect might be due to other mechanism than antioxidative activity [Table 1]. The results were found to be in line with a separate study by Wangensteen *et al*.[3] The latter study also revealed the potent cytotoxic effect and brine shrimp lethality of the lipophilic extracts, which could be attributed to the high amount of tephrosin and other rotenoids [Table 1]. No activity was observed by Alamgir *et al*.[23] when *S. globosus* was tested for neuropharmacological effect on the mice. In a recent study by Kuddus *et al*.[54] the bark extract of the species of Bangladeshi origin exhibited significant cytotoxic effect in the brine shrimp lethality bioassay, along with the membrane stabilizing activity using the murine erythrocyte in the hypotonic solution and significant thrombolytic activity in human blood specimen.

Sonneratia caseolaris

S. caseolaris (L.) Engl. (Sonneratiaceae), locally known as Chak Keora in Bengali, is an evergreen, medium to tall tree growing upto a height of 10 m.[2] Fruits from the species are used to treat bleeding, hemorrhages, piles, sprain poultices.[7] Study by Sadhu *et al*.[55] revealed the presence of two flavonoids [Figure 3], luteolin (39) and luteolin 7-O-β-glucoside (40) possessing antioxidant activity (DPPH radical scavenging activity on TLC) [Table 1] from the dried powdered leaf of the species from Sundarban mangroves of Bangladesh. Ahmed *et al*.[56] observed significant dose dependent effect on the serum glucose and lipid profiles in rats when administered with the dried leaf powder from this species to their diet. The administration had led to the significant decrease in serum glucose, serum triglyceride, serum total cholesterol and serum low density lipoprotein cholesterol levels when compared to controls. The use of leaf powder in the diets had also resulted in significant increases in the serum high density lipoprotein cholesterol levels. The study suggests *S. caseolaris* leaf as a potential source for antidiabetic agents and cure for coronary diseases. A study by Chaudhuri and Guha[20] revealed the ethanolic extract of the leaf of *S. caseolaris* to possess antifungal activity against *F. oxysporum*. In a recent study on the species from Bangladeshi Sundarban, Mubassara *et al*.,[57] showed the species to possess strong antioxidative activity and reducing power. The study also exhibited inhibition of both histamine and leukotriene B_4 suggesting the species to be a good source for the development of anti-allergic agents.

Acanthus ilicifolius

A. ilicifolius L.(Acanthaceae), locally called Horkoch Kanta or Horgoja in Bengali, is a scarcely woody, bushy, dense vine shrub of height upto 2.5 m.[2] Parts of the species are used as aphrodisiac and relief for asthma, diabetes, diuretic, dyspepsia, hepatitis, leprosy, rheumatism and a

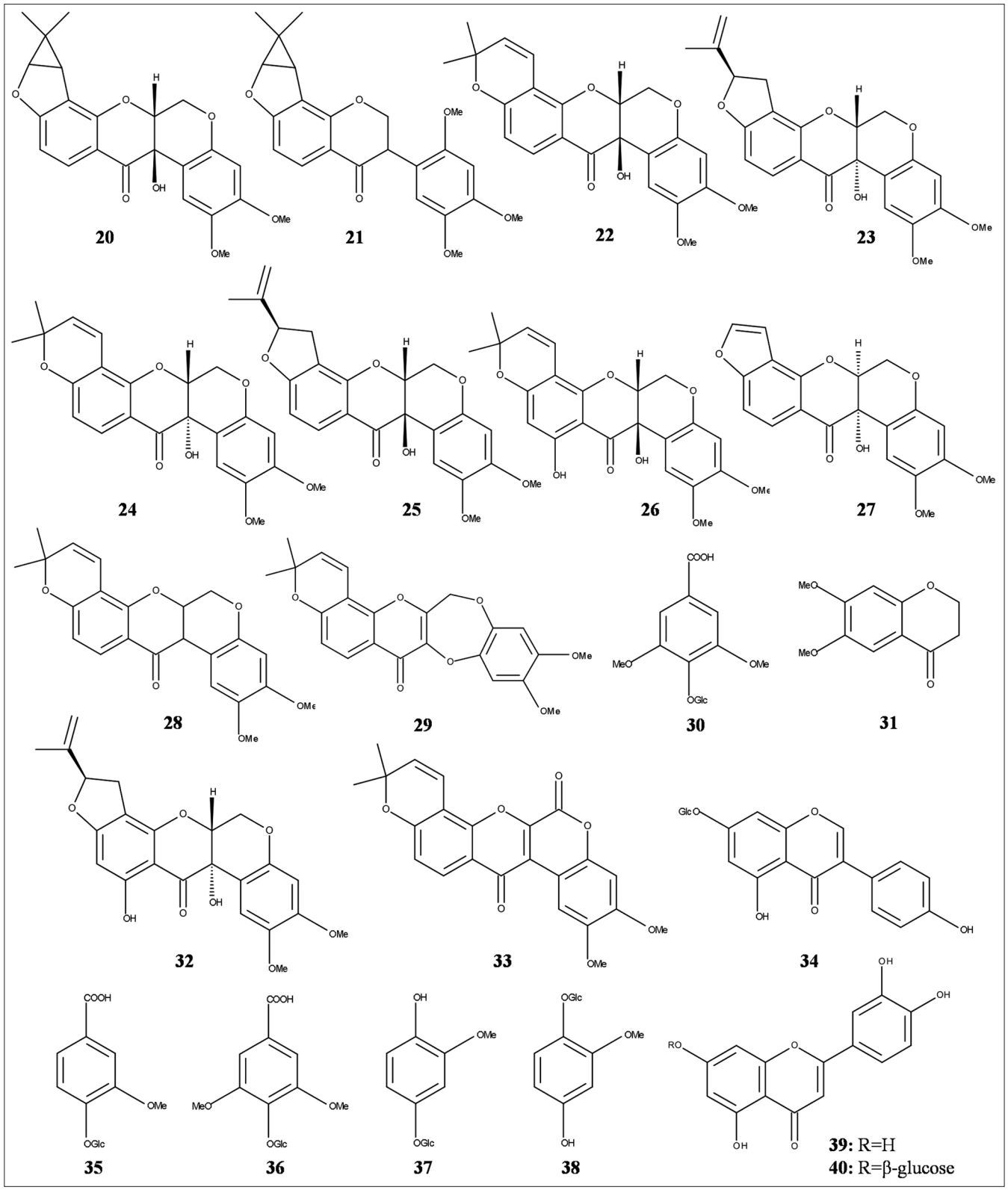

Figure 3: Structures of compounds isolated from *Sarcolobus globosus* (20-38) and *Sonneratia caseolaris* (39,40)

number of ailments.[7] Chakraborty *et al*.[58] showed the chemopreventive potential of the aqueous leaf extract of the species in transplantable Ehrlich ascites carcinoma-bearing murine model manifested in limiting metallothionein protein expression and in preventing DNA alternations in the animal liver. The study demonstrated decreases

in tumor cell count, increase in mean survival duration of the animals, restoration in hematological and hepatic histological profiles. Banerjee *et al*.[18] reported the antioxidant activity of the species from Indian Sundarbans. Senthil Kumar *et al*.[59] demonstrated the anti-inflammatory activity of the leaves of the species from Indian Sundarbans using the murine model. The study showed a considerable decrease of rat paw edema. The fraction of the extract was also found to be a free radical scavenger and is in line with another study by Banerjee *et al*.[18] Leaf and bark extract of the species have been reported to possess activity against a pathogenic fungus, *F. oxysporum*.[20] A recent study by Islam *et al*.[60] on the methanolic extract of *A. ilicifolius* leaves demonstrated dose dependent antinociceptive activity against acetic-acid induced writhing, formalin and hot plate induced murine models. In another report by Senthil Kumar *et al*.,[61] the gastroprotective potential of the methanolic extract of the leaves from the species was demonstrated using different models of gastric ulceration in rats. The extract exhibited protective activity against the aspirin, indomethacin, stress, ethanol and pylorus ligation induced gastric ulcerations.

The information presented in this review clearly indicate that the discussed mangrove species from the Sundarban estuary possess full potential for extraction of pharmacologically vital compounds. Various classes of phytochemicals such as flavonoids, limonoids, rotenoids, phenolic glycosides etc., have been isolated and characterized from these species, a number of which have been reported to possess different types of biological activities. These compounds and their derivatives might be useful in newer drug discovery process. Despite the noticeable biological activities, however, minimum initiatives have been taken for the ethnobotanical and ethnopharmacological studies, search of natural products and establishment of activity-structure relationship. In our literature survey it was noticed that though the species such as *C. decandra*, *E. agallocha*, *A. ilicifolius* exhibited a number of biological activities there's strong lacuna in the search for the active compounds responsible behind these activities. From the Sundarban estuary, among the species discussed, it was found that the majority of the works have been reported on *X. granatum* while *S. caseolaris* was found to be least reported. Attempt to establish activity-structure relationship may also reveal an array of compounds responsible for single or different activities, which might be of synergistic nature. For instance, in a study on *C. decandra*, two of the extracts, which showed significant antimicrobial activity during the disc-diffusion assay failed to exhibit any activity during bioautography after the separation of the compounds using TLC.[21] This might be due to the separation of the constituents, which were showing activity at the synergistic level. Activity guided isolation of bioactive compounds have potent application to establish the activity-structure relationship. The information available also clearly states the lack of intense research regarding the biological activity and the chemical investigation of the aforementioned mangrove species from Indian Sundarban mangrove forest. Majority of the studies on the plants have been reported from Bangladeshi Sundarban estuary. Intense search should be carried out to delve deeper into this sparsely explored promising area from where new bioactive compounds can be isolated eventually helping in the drug discovery process.

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