

Chemical constituents and biological activities of the genus *Subergorgia*

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ABSTRACT

The genus *Subergorgia* (coelenterata, Gorgonacea, Subergorgiidae) is distributed in the Indo-pacific region. Previous investigations on the various species of the genus have revealed the presence of a number of new compounds including alkaloids, sesquiterpenes, diterpenes, and steroids. Certain biological activities particularly cytotoxic activity have been observed for the isolated constituents and compositions derived from the coral. This review covers the secondary metabolites reported from the genus *Subergorgia* and their biological properties.

Key words: Alkaloids, cytotoxic activity, steroids, *Subergorgia*, terpenes

INTRODUCTION

The family Subergorgiidae comprises calcareous sclerites corals; members of the genus *Subergorgia* possess fan- or brush-like branches. *Subergorgia suberosa* [Figure 1] is one of the most widely distributed species particularly found in the Indo-pacific tropical regions from the Red Sea to the Central Pacific. It is mostly found attached to reef slopes at depths of 15–20 m, and in terms of size it is usually 50 cm high and wide. A wide range of new secondary metabolites have been isolated from the genus *Subergorgia* comprising alkaloids, sesquiterpenes, diterpenes, and steroids.^[1-10]

Taxonomy

Kingdom: *Animalia*

Phylum: *Cnidaria*

Class: *Anthozoa*

Order: *Alcyonacea*

Family: *Subergorgiidae*

Genus: *Subergorgia*

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Morphology

S. suberosa possess white tentacles, reddish brown polyps and small spindle-like sclerites. The other species of the genus *Subergorgia* possess free branches and densely branched fans. The sclerites vary in colour from yellowish red to dark red and even colour less in certain species.^[3]

CHEMICAL CONSTITUENTS AND BIOLOGICAL ACTIVITIES

The chemicals constituents isolated from the various species of the genus *Subergorgia* are detailed below and the new compounds reported are listed in Table 1.

Subergorgia hicksoni

Chromatographic separations of *S. hicksoni* led to the isolation of

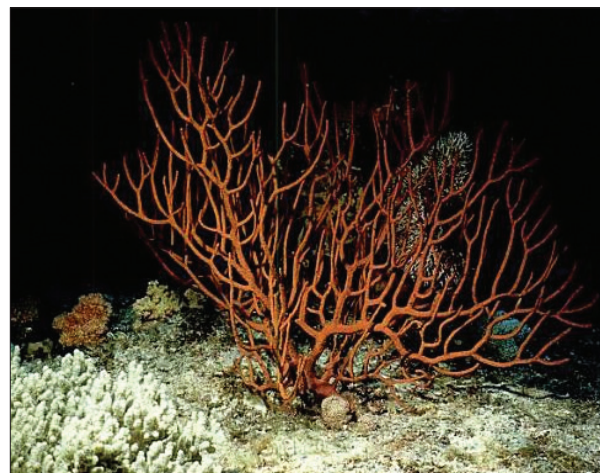


Figure 1: *Subergorgia suberosa*

Table 1: New chemical constituents from the genus *Subergorgia*

Compound	No.	Class	Species	Reference
6-(1'-purine-6', 8'-dionyl)-suberosanone	30	Alkaloid	<i>S. suberosa</i>	[30]
3, 9-(2-imino-1-methyl-4-imidazolidinone-5-yl) isopropenylpurine-6, 8-dione	31	Alkaloid	<i>S. suberosa</i>	[30]
1-(3'-carbonylbutyl) purine-6, 8-dione	32	Alkaloid	<i>S. suberosa</i>	[30]
9-(3'-carbonylbutyl) purine-6, 8-dione	33	Alkaloid	<i>S. suberosa</i>	[30]
4-carboxy-5, 6-dihydro-4H, 8H-pyrimido [1, 2, 3-cd] purine-8, 10(9H)-dione	34	Alkaloid	<i>S. suberosa</i>	[8]
7, 9-dihydro-1-(3-oxobutyl)-1H-purine-6, 8-dione	35	Alkaloid	<i>S. suberosa</i>	[8]
7-hydro-9-(3-oxobutyl)-1H-purine-6, 8-dione	36	Alkaloid	<i>S. suberosa</i>	[8]
Reticulolide	6	Diterpene	<i>S. reticulata</i>	[14]
(+)-(7R, 10S)-2-methoxy calamenene	9	Sesquiterpene	<i>S. reticulata</i>	[6]
(+)-(7R, 10S)-2-methoxy-5-acetoxy calamenene	8	Sesquiterpene	<i>S. reticulata</i>	[6]
2 β -acetoxysubergorgic acid	25	Sesquiterpene	<i>S. suberosa</i>	[23]
2 β -hydroxysubergorgic acid	15	Sesquiterpene	<i>S. suberosa</i>	[7]
5-hydroxy-8-methoxycalamenene	2	Sesquiterpene	<i>S. hicksoni</i>	[5]
6-(9'-purine-6', 8'-diolyl)-2 β -suberosanone	26	Sesquiterpene	<i>S. suberosa</i>	[25]
8-methoxy-methoxycalamenene	1	Sesquiterpene	<i>S. hicksoni</i>	[5]
Methyl 2 β -acetoxysubergorgate	14	Sesquiterpene	<i>S. suberosa</i>	[7]
Methyl 2 β -hydroxysubergorgate	13	Sesquiterpene	<i>S. suberosa</i>	[7]
Methyl subergorgate	16	Sesquiterpene	<i>S. suberosa</i>	[7]
Sesquiterpenes (+)-(7R, 10S)-2, 5-dimethoxy calamenene	7	Sesquiterpene	<i>S. reticulata</i>	[6]
Subergorgic acid	10	Sesquiterpene	<i>S. suberosa</i>	[16]
Subergorgiol	24	Sesquiterpene	<i>S. suberosa</i>	[23]
Suberosenone	11	Sesquiterpene	<i>S. suberosa</i>	[17]
Suberosol A	20	Sesquiterpene	<i>S. suberosa</i>	[22]
Suberosol B	21	Sesquiterpene	<i>S. suberosa</i>	[22]
Suberosol C	22	Sesquiterpene	<i>S. suberosa</i>	[22]
Suberosol D	23	Sesquiterpene	<i>S. suberosa</i>	[22]
(22E)-14 α -hydroxy-cholesta-1, 4, 22-trien-3-one	37	Steroid	<i>S. suberosa</i>	[33]
11 α , 15 α -diacetoxy-17 α -pregna-4, 20-dien-3-one	3	Steroid	<i>S. mollis</i>	[11]
24-methylcholest-7, 22 E-diene-3 β , 5 α , 6 β , 25-tetraol	29	Steroid	<i>S. suberosa</i>	[9]
24R-methyl-3 β , 6 α , 11-trihydroxy-9, 11-seco-5 α -cholest-7, 22E-diene-9-one	19	Steroid	<i>S. suberosa</i>	[19]
24S-methyl-3 β , 6 α , 11-trihydroxy-9, 11-seco-5 α -cholest-7, 22E-diene-9-one	18	Steroid	<i>S. suberosa</i>	[19]
3-(1', 2'-ethandiol)-cholest-3 β , 5 α , 6 α , 11 α -tetraol	38	Steroid	<i>S. suberosa</i>	[33]
3-(1', 2'-ethandiol)-24- methylcholest-8(9), 22E-diene-3 β , 5 α , 6 α , 7 α , 11 α -pentaol	28	Steroid	<i>S. suberosa</i>	[9]
3, 9-dioxo-9, 11-secocholesta-5, 7-dien-11-al	12	Steroid	<i>S. suberosa</i>	[1]
3 β , 6 α , 11, 20 β , 24-pentahydroxy- 9, 11-seco-5 α -24-ethylcholest-7, 28-diene-9-one	27	Steroid	<i>S. suberosa</i>	[9]
3 β , 6 α , 11-trihydroxy-9, 11-seco-5 α -cholest-7-ene-9-one	17	Steroid	<i>S. suberosa</i>	[19]
Reticulatic	4	Steroid	<i>S. suberosa</i>	[10]
Reticulatin	5	Steroid	<i>S. suberosa</i>	[10]

two new [Figure 2a] sesquiterpenes 8-methoxy-methoxycalamenene (1) and 5-hydroxy-8-methoxycalamenene (2).^[5]

Subergorgia mollis

Wu *et al.* reported a new steroid [Figure 2a], 11 α , 15 α -diacetoxy-17 α -pregna-4, 20-dien-3-one (3), along with a known steroid 17 α -pregna-4, 20-dien-3-one, from the formosan gorgonian *S. mollis*.^[11]

Subergorgia reticulata

Guo reported five known polyhydroxylated steroids anyaols A-E [Figure 2a], among which some exhibited cytotoxic activity.^[12] Investigations of the dichloromethane/ethanol extract of the South China Sea gorgonian coral *S. reticulata* resulted in the

identification of two new compounds reticulatic acid (4) and reticulatin (5) and a known compound 3, 22, 25-trihydroxy-16-24, 20-24-bisepoxy-3 β , 16 β , 20S, 22R, 24S-cholest-5-ene.^[10]

Yang *et al.* isolated nine known compounds batyl alcohol, cholesterol, cholesta-7, 22-diene-3 β , 5 α , 6 β -triol, ergostra-7, 22-diene-3 β , 5 α , 6 β -triol, 5, 8-epidioxycampesta-6, 22-dien-3-ol, guanine, theine, thymine, and uracil from the South China Sea gorgonian coral *S. reticulata*.^[13]

Isolations of the ethyl acetate extract of the Chinese gorgonian coral *S. reticulata* afforded a new briarane-type diterpenoid reticulolide (6), together with the known compounds (-)-11 α , 20 α -epoxy-4-deacetyljunceollolide D, junceollin, junceollolide A,

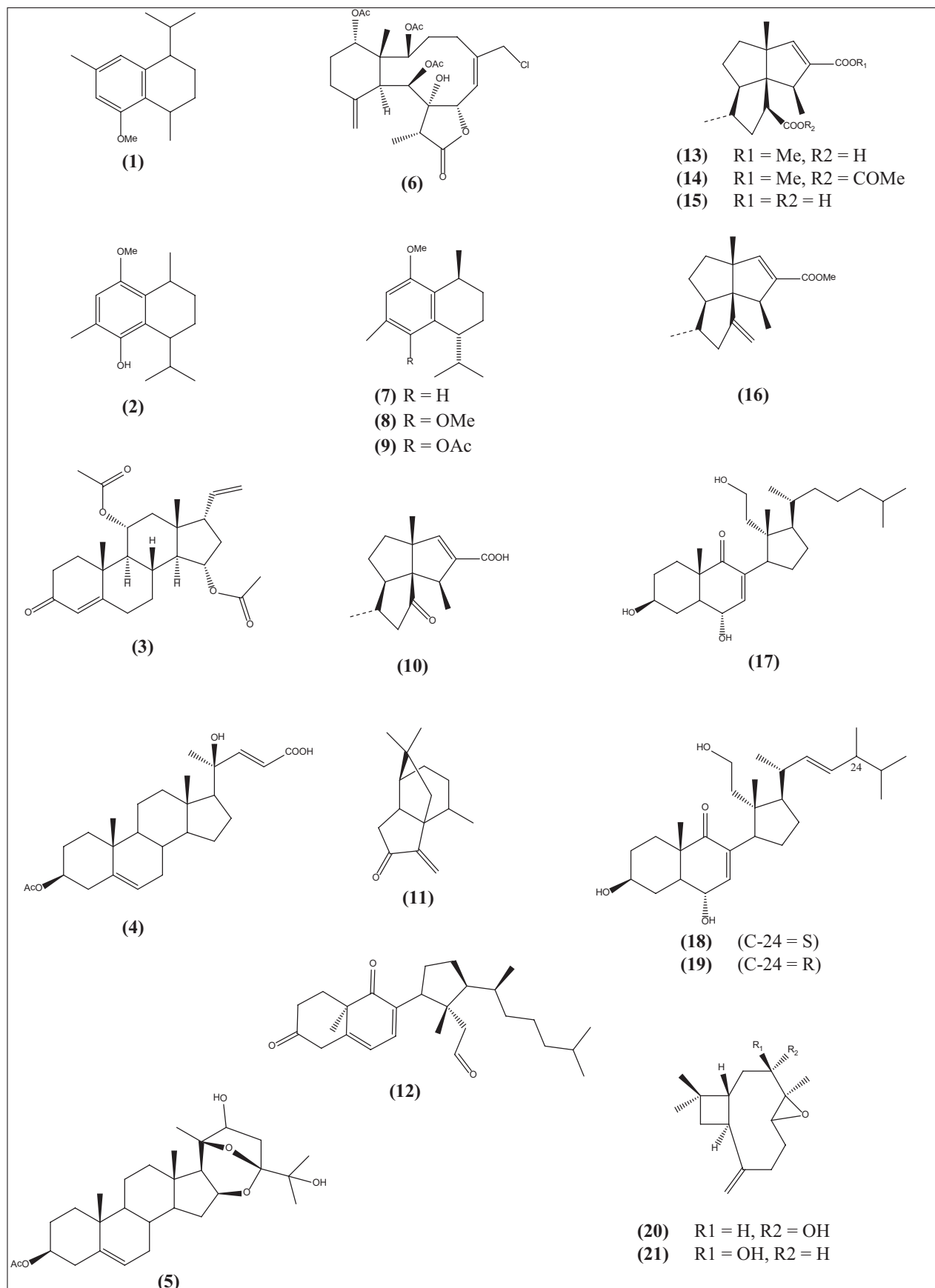


Figure 2a: Constituents (1-21)

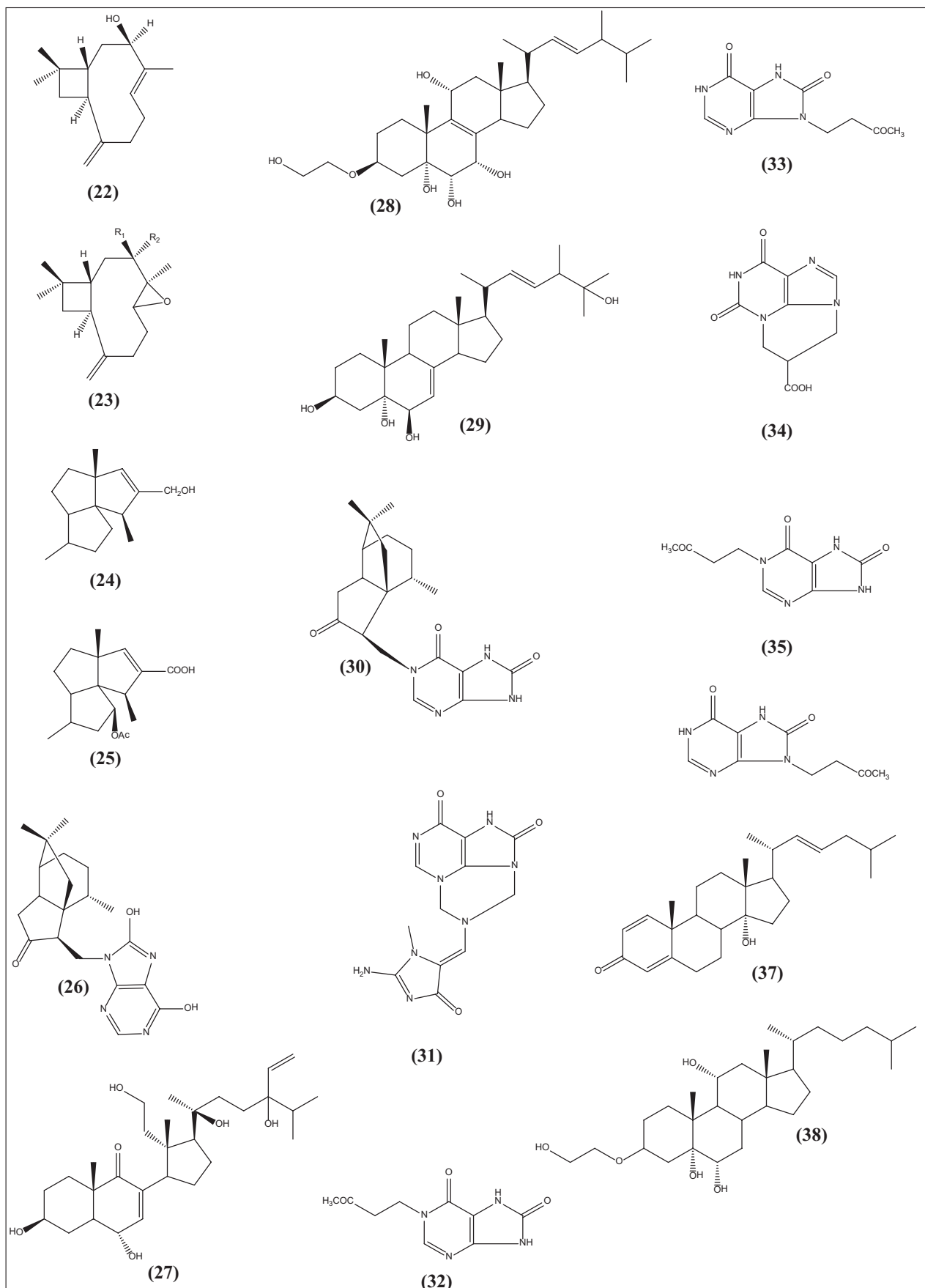


Figure 2b: Constituents (22-38)

praelolide, umbraculolide A, and umbraculolide C. Among these compounds, praelolide and junceellin exhibited anti-settlement activity against the larva of *Bugula neritina* at a concentration of 50 µg/mL.^[14]

Limna Mol *et al.* isolated three new sesquiterpenes (+)-(7R, 10S)-2,5-dimethoxy calamenene (7), (+)-(7R, 10S)-2-methoxy-5-acetoxy calamenene (8), and (+)-(7R, 10S)-2-methoxy calamenene (9) from the methanol extract of the Indian gorgonian *S. reticulata*. The isolated compounds (7-9) showed potent inhibitory effects against cyprids of *Balanus amphitrite* with EC₅₀ values of 4.4, 7.8, and 0.03 µg/mL, respectively. The compounds also showed appreciable activity against *Artemia nauplii* with an EC₅₀ value 50 µg/mL.^[6]

Bioguided isolations of *S. reticulata* resulted in the identification of three known compounds (+)-(7R, 10S)-2-methoxy calamenene, (+)-(7R, 10S)-2,5-dimethoxy calamenene, and (+)-(7R, 10S)-2-methoxy,5-acetoxy calamenene. Among these compounds, (+)-(7R, 10S)-2-methoxy,5-acetoxy calamenene showed potent antifouling activity against the cyprids of *B. amphitrite* with EC₅₀ value of 0.0335 µg/mL and a high therapeutic ratio of 799.^[15]

Subergorgia suberosa

Isolations of the gorgonian coral *S. Suberosa* led to the isolation subergorgic acid (10) [Figure 2a and 2b], which showed cardiotoxic properties and significant activity against “Soman” toxicity in mice.^[16-17] Bokesch *et al.* reported a new cytotoxic sesquiterpene, suberosenone (11), along with known piscicidal sesquiterpenes, buddledins C-D, suberosenone indicated significant cytotoxic activity against tumor cell lines.^[18]

A new steroid 3,9-dioxo-9,11-secocholesta-5,7-dien-11-al (12) was reported from *S. suberosa* together with the known compounds avenasterol, campesterol, cholesterol, fucosterol, 3β-hydroxypregn-5-en-20-one, 24-propylidenecholest-5-en-3β-ol, subergorgic acid, lathosterol, and zymosterol.^[1]

Chemical investigation of the methanol extract of the gorgonian coral *S. suberosa* obtained from the Indian ocean resulted in isolation of four new compounds, methyl 2β-hydroxysubergorgate (13), methyl 2β-acetoxysubergorgate (14), 2β-hydroxysubergorgic acid (15), and methyl subergorgate (16), along with the known compound subergorgic acid.^[7]

Chromatographic separations of the *S. suberosa* afforded three new secosterols, 3β, 6α, 11-trihydroxy-9, 11-seco-5α-cholest-7-ene-9-one (17), 24S- methyl-3β, 6α, 11-trihydroxy-9, 11-seco-5α-cholest-7, 22E-diene-9-one (18), and 24R-methyl-3β,6α,11-trihydroxy-9, 11-seco-5α-cholest-7, 22E-diene-9-one (19).^[19] A compound Subergorgia suberosato sesquiterpene was isolated from *S. suberosa* which is suggested to act as a new antidote for soman. A procedure was developed for isolating isosubergorgic acid from *S. suberosa*; isosubergorgic acid is claimed to possess acetylcholine esterase inhibitory activity, thus it is suggested to treat Alzheimer's disease.^[20-21]

Chromatographic separations of the Taiwanese gorgonian coral *S. suberosa* resulted in the identification of four new β-caryophyllene-derived sesquiterpenes alcohols, suberosols A-D (20-23), together with two known β-caryophyllene-derived sesquiterpenes ketones, buddledins C-D. Certain isolated constituents exhibited cytotoxic activity.^[22]

Chromatographic separations of the ethyl acetate extract of the Taiwanese Gorgonian coral *S. suberosa* led to the isolation of two new subergane-based sesquiterpenes, subergorgiol (24) and 2β-acetoxysubergorgic acid (25), together with four known compounds subergorgic acid methyl ester, subergorgic acid 2β-acetoxy methyl ester, 2β-hydroxysubergorgic acid, and subergorgic acid. Among the compounds, subergorgic acid methyl ester showed moderate cytotoxic activity against the growth of HeLa cancer cells.^[23] Subrahmanyam *et al.* reported 3,3-dimethoxy-5α-pregnan-20-one gorgonian *S. suberosa*.^[24]

Chromatographic separations of the ethanol/dichloromethane extracts of the South China Sea gorgonian *S. suberosa* yielded a new sesquiterpene alkaloid, 6-(9'-purine-6',8'-diolyl)-2β-suberosanone (26), along with three known compounds, suberosenol A, subergorgic acid, and subergorgiol. Among the compounds, (26) indicated moderate cytotoxic activity against the human breast carcinoma MDA-MB-231 cell line with an IC₅₀ value of 8.87 µg/mL.^[25]

Examinations of the gorgonian sea coral *S. suberosa* collected from the Mandapam coast, resulted in the identification of four compounds, batyl alcohol, subergorgic acid N-hexadecanoyl-2-amino-1,3-dihydroxyoctadec-4-ene, and thymine.^[26] A formulation was developed for the isolation of a sesquiterpene ketone, which is claimed to inhibit the growth of tumor cells and treat breast cancer, leukemia, oral cancer, liver cancer, and lung cancer.^[27]

Isolations of the ethanol/dichloromethane extract of the South China Sea gorgonian *S. suberosa* afforded three new polyhydroxylated sterols, 3β,6α,11,20β,24-pentahydroxy-9,11-seco-5α-24-ethylcholest-7,28-diene-9-one (27), 3-(1',2'-ethandiol)-24-methylcholest-8(9),22E-diene-3β,5α,6α,7α,11α-pentaol (28), and 24-methylcholest-7,22 E-diene-3β,5α,6β,25-tetraol (29) along with six known steroids 24α-methylcholest-7,22E-diene-3β,5α,6β,9α-tetraol, 24α-methylcholest-7,22-dien-3β,5α,6β-triol, 3β,6α,11-trihydroxy-9,11-seco-5α-cholest-7-ene-9-one, 3β,6α,5,11-tetrahydroxy-9,11-seco-5α-cholest-7-ene-9-one, 3-O-β-D-glucopyranosyl-β-sitosterol, and 25-O-acetyl-3-O-[β-D-arabinopyranosyl-oxy]-cholest-5-ene-3β,19,25-triol.^[9]

A steroid was isolate a steroid 3β,6α,11,20β,24-pentahydroxy-9,11-seco-5α-24-ethylcholest-7,28-diene-9-one from *S. suberosa*. The constituent is claimed to inhibit the growth of cancer cells of gastric cancer, leukemia, and liver cancer.^[28] A purine alkaloid, 3,9-(2-imino-1-methyl-4-imidazolidinone-5-yl)-isopropenyl-purine-6,8-dione, was isolated from *S. suberosa*. The isolated alkaloid is claimed to inhibit the growth of cancer

cells of breast adenocarcinoma, liver cancer, gastric cancer, and leukemia.^[29]

Isolations of the ethanol/dichloromethane extracts of the South China Sea gorgonian *S. suberosa* led to the isolation of four new purine alkaloids, 6-(1'-purine-6',8'-dionyl)suberosanone (30), 3,9-(2-imino-1-methyl-4-imidazolidinone-5-yl)isopropenylpurine-6,8-dione (31), 1-(3'-carbonylbutyl)purine-6,8-dione (32), and 9-(3'-carbonylbutyl)purine-6,8-dione (33), along with three known compounds, guanosine, thymidine, and adenosine. The compounds (30-33) indicated weak cytotoxic activity against human cancer cell lines MDA-MB-231 and A435.^[30]

Bioguided isolations of *S. suberosa* led to the isolation of six known compounds, subergorgic acid, pregn-4-ene-3, 20-dione (progesterone), 5 β -pregn-3, 20-dione, 3 β -pregn-5-ene-20-one-3-ol, 3 β ,5 β -pregn-20-one-3-ol, and stigma-7,22-dien-3 β ,5 α ,6 β -triol. The isolated constituents were antilarval against *B. amphitrite* and *B. neritina* larvae and antibacterial against 15 marine bacterial strains. Among the constituents, pregn-4-ene-3, 20-dione showed the most potent activity.^[31]

Chromatographic separations of the dichloromethane/methanol extract of the South China Sea gorgonian coral *S. suberosa* led to the isolation of two new steroids, 3 β -O-palmitoyl-pregn-5-en-20-one-3-ol (1) and 3 β -O-palmitoyl-5 α -pregn-20-one-3-ol (2), along with six known steroids, 5 α -pregn-1-ene-3,20-dione, 3 β , 5 α -pregn-20-on-3-ol, 3 β -pregn-5-en-20-on-3-ol, 3 β , 5 β -pregn-20-on-3-ol, 5 β -pregn-3, 20-dione, and pregn-4-en-3, 20-dione. Furthermore, this is the first report of these compounds from this coral.^[32]

Qi et al. reported three new purine derivatives, 4-carboxy-5, 6-dihydro-4H, 8H-pyrimido[1, 2, 3-cd]purine-8, 10(9H)-dione (34), 7, 9-dihydro-1-(3-oxobutyl)-1H-purine-6, 8-dione (35), and 7-hydro-9-(3-oxobutyl)-1H-purine-6, 8-dione (36), from the ethanol/dichloromethane extracts of the South China Sea gorgonian *S. suberosa*.^[8] Two new steroids, (22E)-14 α -hydroxycholesta-1, 4, 22-trien-3-one (37) and 3-(1', 2'-ethandiol)-cholest-3 β , 5 α , 6 α , 11 α -tetraol (38), were reported from *S. suberosa*. The isolated compounds showed cytotoxic activity against human cancer cell lines A549, HONE1, and HeLa. The compounds also indicated antilarval activity against *B. amphitrite* and *B. neritina* larvae.^[33]

Examinations of the ethyl acetate extract of gorgonian *S. suberosa* afforded nine known compounds, cholesta-5-ene-3 β ,7 α -diol, cholestane-1 β ,3 β ,5 α ,6 β -tetrol, cholesterol, (E)-N-2-(1,3-dihydroxy octadecan-4-en)-hexadecamide, batyl alcohol, thymidine, thymine, uracil, and heptadecane. However, this was the first report of these compounds from *S. suberosa*.^[34] Reddy et al. isolated the known compounds subergorgic acid and subergorgic acid methyl ester from *S. suberosa*.^[35]

Subergorgia sp.

Bioguided isolations of *Subergorgia* sp. led to the isolation of

astaxanthin; the isolated constituent showed significant cytotoxic activity against human leukemia cell line K562 and inhibited both TNF- α -induced NF- κ B-DNA binding and TNF- α -induced I κ B α degradation, and nuclear translocation of p50/p65.^[36]

CONCLUSION

A significant number of phytopharmacological investigations on the genus *Subergorgia* show that it is a valuable source of new compounds. However, a limited number of investigations have been attempted on certain species such as *S. bicksoni* and *S. mollis*, hence it would be valuable to conduct bioguided phytochemical studies on these species for isolating new secondary metabolites that could possess cytotoxic or other important biological properties.

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