A Concise Review on Medicinally Important Phytochemical Class: Meroterpene

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Abstract

Meroterpenes are promising drug compounds isolated from microorganisms, marine invertebrates, and higher plants, having a partial terpenoid skeleton and obtained from both terpenoid and polyketide biogenesis. Among the organisms of marine origin that are a rich source of structurally varied meroterpenes, marine fungi and brown algae are promising sources. Meroterpenes are complex molecules with unique molecular architectures derived from intra- and inter-molecular cyclisation or rearrangements and found to exert potent bioactivities in all the domains of life. Important compounds like ubiquinone-10, α-tocopherol, and austinol are included in the meroterpenoid class. Meroterpenoids exhibit antineoplastic, antibiotic, antifungal, antimalarial, analgesic and anti-inflammatory effect. A few compounds of the meroterpenoids category include antidepressant - hyperforin, psychoactive drug - tetrahydrocannabinol, and immunosuppressant - mycophenolic acid. This review aims to provide a detailed overview of meroterpenes along with their source, structures, detailed pharmacological potential, and biological synthesis that we hope will underpin future work on this class of secondary metabolites.

Keywords: austinol, meroterpenes, polyketide biogenesis, psychoactive drug, ubiquinone-10

1. Introduction

Meroterpenes belong to the class of terpenoids mainly found to occur in terrestrial animals, plants, bacteria, fungi, lichens, and marine organisms, which are involved in the electron transport processes and photosynthesis(1). The prefix mero is derived from the Greek *meros*, meaning “part,” “partial,” or “fragment”) is used to describe several secondary metabolites known as meroterpenoids(2). The term m`eroterpenoid was first applied by Cornforth in 1968 to describe the natural products of mixed biosynthetic origin that are derived via the acetate-mevalonate pathway from terpenoid precursors, as well as from polyketide biogenesis(2). Organisms, particularly of fungal origin, show structural diversity and abundance, possibly due to their capability to explore this route of biosynthesis for complex metabolite production. This group of compounds displays potent pharmacological actions such

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as anti-inflammatory[3–6], antioxidant[7,8], antitumor[9–12], anti-viral[6] and antibacterial[13–15] activity.

Meroterpenoids are primarily classified according to their biosynthetic origin, with two main categories identified: polyketide-terpenoids and non-polyketide-terpenoids. Meroterpene biosynthesis enhances the structural diversity beyond that produced by isoprenoid pathways alone. This results in compounds with a wide range of chemical structures and biological activities, driven by variations in the non-terpenoid moiety, the length of the terpenoid chain, the cyclization pattern of the terpenoid portion, and subsequent tailoring reactions[16]. The non-terpenoid part could be polyketides, phenols, alkaloids, and amino acids. The availability of the compounds depends on the source and climate. Among all types of organisms that produce meroterpenoids, fungi are a vibrant source for this group of natural products, and several clinically used drugs or promising leads are obtained, as exemplified by the immunosuppressant agent mycophenolic acid[17] and the antimicrobial agent fumagillin[18]. Recent studies focused on uncovering the molecular bases for the biosynthesis of fungal meroterpenoids have revealed many enzymes responsible for chemical transformations. The review discusses several important compounds that have been isolated and synthesised till now. The information collected here includes sources, chemistry, biosynthesis, and pharmacological activities of meroterpenoids obtained from different natural sources is a valuable resource for developing new drug leads.

2. Sources

Meroterpenoids are found to occur in the marine organisms such as algae, sponges, fungi, etc [1][19]. About 460-470 new terpenes are reported from the wide range of marine fungi[20]. Other than marine sources, some of the plants have been reported for their presence, such as *Psidium guajava* [21], *Psoralea corylifolia* [22], *Rumex madaio* [23] *Rhododendron dauricum*, *Eucalyptus globulus* fruits, *Eucalyptus robusta*, *Baeckea frutescens*, *Hypericum japonicum*. A detailed description of sources is discussed in Table 1.

Table 1: Sources of meroterpene

Class of Source	Source	Compound	Reference
Alga-derived fungi	*Alternaria alternata* k21-1	Tricycloalterfurenes A-D and TCA-F	(121)
Antarctic fungus	*Aspergillus terreus*	terreustoxins A - M	(110)
Arctic fungi	Eutepella species D-1	Eutypellacytosporins A, Eutypellacytosporins B, Eutypellacytosporins C, Eutypellacytosporins D	(122)
Coral-assocd. Fungi	Alternaria sp. ZH-15	Alternarin A, Macrophorins A, and Macrophorins B	(86)

Crinoid-derived Fungi	*Stachybotrys chartarum* 952	Stachybonoids A–F	(6)
Deep-sea-derived fungi	Penicillium species YPGA11	Andrastin A, 3-deacetylcitreohybridonol, citreohybridonol, andrastin B, 6α-hydroxyandrastin B, dihydrocitreohybridonol, conidiogenone C, conidiogenone I	(111)
	Penicillium allii-sativi	Andrastone A	(112)
	Phomopsis tersa FS441	Photeroids A, Photeroids B	(103)
Endophytic fungal	*Lycium barbarum*	Bipolahydroquinones A-C	(123)
	Pseudocosmospora species Bm-1-1	Cosmosporin A, 8'-hydroxy-cannabiorcichromenic acid, cannabiorcichromenic acid, and decarboxycannabiorcichromenic acid	(120)
	Penicillium sp. sh18	Isopenicins A-C	(124)
Marine fungi	Talaromyces species CX11	Talaromyolides A−D and talaromytin	(72)
	Penicillium species SF-5497	Preaustinoid A6 & Preaustinoid A7	(105)
	Aspergillus terrus GZU-31-1	Furanasperterterpenes A, Furanasperterterpenes B, and 11-acetoxy-terretonin E	(11)
	Aspergillus versicolor	Asperversins A-G	(89)
	di-Et sulfate (DES) mutant *Penicillium chrysogenum* S-3-25	Chrysomutanin	(10)
	Penicillium rudallense	Austalides V, Austalides W, Austalides X	(102)
	Antrodia cinnamomea	Antroquinonols N-X	(118)
	Ganoderma cochlear	Cochlearols E-M Gancochlearols	(125)(3) (126)

Ganoderma fungus (fruiting body and mushroom)		(±)-cochlearins A-E, F-I	
	Ganoderma theaecolum	Ganotheaecoloids A-N	(4)
	Ganoderma resinaceum	Ganoresinains A–E	(127)
	Ganoderma applanatum	Applanatumines B-D	(78)
	Ganoderma leucocontextum	Spiroapplanatumine K, 6 spiroapplanatumine L, 6- (±)-spirolingzhine A and spirolingzhine D	(76)
Genus Emericella (Ascomycota)	*E. variecolor*	varitriol, varioxirane and dihydroterrein, Emervaridione and varioxiranediol	(128)
	Emericella sp. HK-ZJ	Austin, austinol, dehydroaustin and acetoxydehydroaustin	
Jellyfish-derived fungi	*P. chrysogenum* J08NF-4	Chrysogenester	(5)
Mangrove endophytic fungus	*Aspergillus versicolor*	(7R,8R)-8- hydroxysydowic acid, (7S,10S)-10-hydroxy-sydowic acid, (7S,11R)-12-hydroxy-sydowic acid, (7S,11R)-12- acetoxy-sydowic acid, and (7R,8R)-1,8-epoxy-11-hydroxy-sydonic acid 9	(129)
	Phyllosticta capitalensis	Guignardone A, 12-hydroxylated guignardone A, Guignardone J, Guignardone M	(14)
	Diaporthe species SCSIO 41011	Chrodrimanins A, B, E-H	(130)
	Penicillium simplicissimum MA-332	Simpterpenoid A	(8)
	Penicillium species TGM112	Penicianstinoids A & B	(15)
Mariana Trench sediment-derived fungi	Penicillium species SY-2107	Andrastones B, Andrastones C	(113)

Marine sediment-derived	Aspergillus flocculosus	12-epi-aspertetranone D	(12)
Sipunculus nudus, Marine worm	Penicillium species SCS-KFD09	Chrodrimanins O-S	(101)
Plant pathogenic fungus	Verticillium albo-atrum	Acetoxydehydroaustin A and austin	(104)
Thermophilic fungus	Aspergillus terreus TM8	Terretonin M and A	(131)
Wrinkled peach mushroom	Basidiomycete Rhodotus palmatus	Rhodatin	(7)

3. Biosynthesis

Meroterpenes are generally synthesized via the aceto-mevalonic acid pathway, but sometimes it may be synthesized via the methylerythritol phosphate (MEP) pathway, as it is responsible for the synthesis of isopentenyl diphosphate (IPP), which is the primer unit required for further synthesis[24,25].

4. Phytochemistry of Meroterpenes

4.1 Terrestrial Plants and Fungi Meroterpenes

Meroterpene is a part of terpene; hence, the basic structural moiety is similar to terpene, it contains units of isoprene in a head-to-tail style. These arrangements can be varied by synthetic pathway. Cyclisation of the isoprene unit is majorly seen in identifying compounds that can be monocyclic, bicyclic, tricyclic, and so on[20].

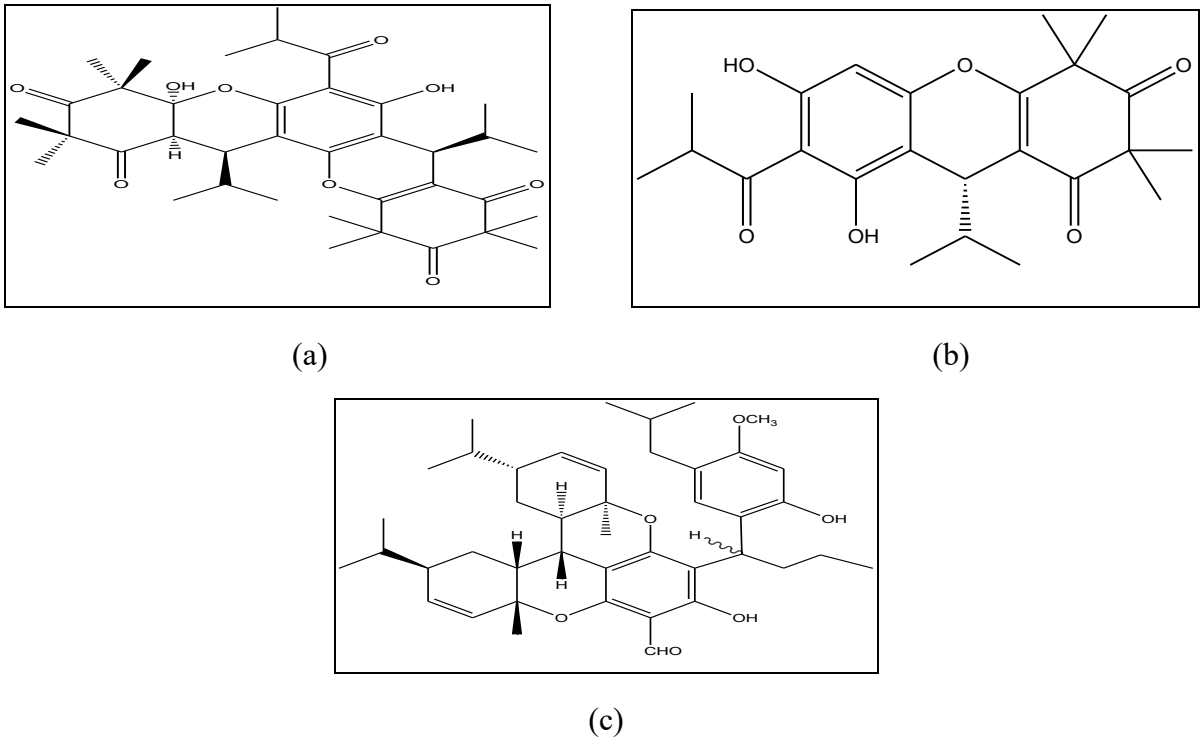
4.1.1 Phloroglucinol-Based Meroterpenoids

4.1.1.1 Dimeric Phloroglucinol-Phellandrene Meroterpenoids

Eucalyptusdimers A-C are reported as dimeric phellandrene-derived meroterpenoids obtained from the fruits of *Eucalyptus robusta*. They have a unique fused skeleton between 2 phellandrenes & 2 phloroglucinol subunits[26]. Glabralides A-C were obtained from the entire plant of *Sarcandra glabra,* where glabralide A has a novel Diels-Alder adduct of the chalcone and α-phellandrene, consisting of a unique bicyclo[2.2.2]octene core unit with five chiral centers, and glabralide C was discovered as an unusual α-phellandrene cyclo-adduct with o-hydroxy phenylacetate[27], Callisalignones A-C, callisalignenes A-F, myrtucommulone D (Fig. 1a), and isomyrtucommulone B (Fig. 1 b) were obtained from the twig and leaf of *Callistemon salignus*. Callisalignones B and C have an adduct of β-triketone and acylphloroglucinol, while callisalignenes A-D have acylphloroglucinol and α-phellandrene with different coupling models via hetero-Diels-Alder reaction[28].

Eucalyptusdimers A (Fig. 1c) is reported for acetylcholinesterase inhibitory activity[26]. Myrtucommulone D consists of an isobutyrophenone core (IBP-C), substituted with two syncarpic acid moieties, and represents an oligomeric, nonprenylated acylphloroglucinol which

is reported from leaves of myrtle (*Myrtus communis* L., Myrtaceae)[29] It is reported to possess potent antibacterial activity on *Staphylococcus aureus* and drug-resistant *Staphylococcus aureus* strains. Isomyrtucommulone B is also reported for antibacterial activity against *Escherichia coli*[28].



(a) (b)

(c)

Fig 1. Various Dimeric Phloroglucinol-Phellandrene Meroterpenoids (a) Myrtucommulone D (b) Isomyrtucommulone B (c) Eucalyptusdimers A

4.1.1.2 Formyl-Phloroglucinol–Terpene Meroterpenoids

Formyl phloroglucinol meroterpenoids are sesquiterpene-based hybrid natural products. Major genus sources of these metabolites are *Psidium, Eucalyptus, Myrtus,* and *Rhodomyrtus*. Known pharmacological activities of this class include cytotoxicity, HIV-RTase inhibitory, PTP1B enzyme inhibitory, and antibacterial activity. Particularly, the cytotoxic effect of this type of class is noticeable[30].

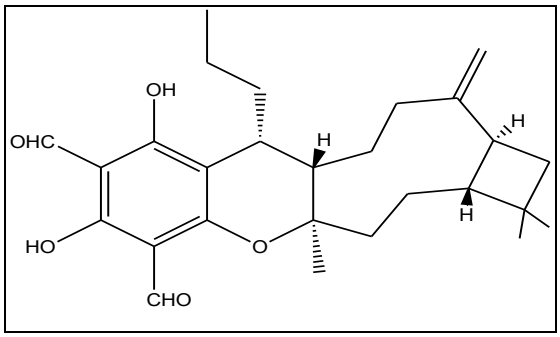
Littordials A–E are meroterpenoids composed of phloroglucinol and β-caryophyllene units, isolated from the leaves of *Psidium littorale*. In their structure, they have a unique acyl phloroglucinol unit where littordial E has a sporadic 6,7,9,4-fused tetracyclic ring system[30]. Littordials B-C and E (Fig. 2a, b & c) are reported to be cytotoxic on MDA-MB-231 & B16 human cancer cell line study[30].

Eucalrobusones A-I are obtained from the leaf of *Eucalyptus robusta*. Eucalrobusones A and B possess a unique structure in which a maaliane-type sesquiterpene moiety is linked to a formyl-substituted position on the phloroglucinol ring. Eucalrobusone E has a chroman ring which is connected with a bicyclogermacrane nucleus at C-3/C-4 position. Eucalrobusone F structure has an aromatic dehydromenthane monoterpene unit which is attached through a formyl-derived carbon atom. Eucalrobusones G-I are unique among all the structures; they

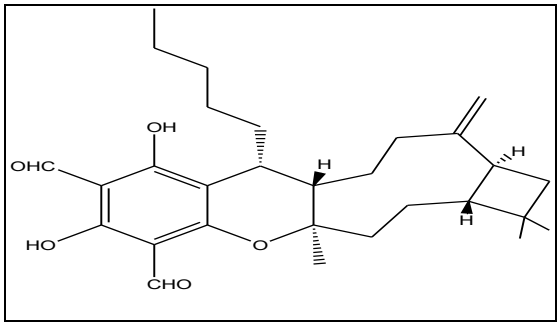
were first discovered as cubebane-based formyl-phloroglucinol meroterpenoids attached with a rare 1-oxaspiro [5.5] undecane core [31][32]. Among eucalrobusones A-I, only eucalrobusone C (Fig. 2d) showed significant cytotoxic activity[31][32].

Eucalypglobulusals A-J reported from the fruits *Eucalyptus globulus*[33]. Eucarobustol E is obtained from the *Eucalyptus robusta*[34]. Eucalypglobulusal F (Fig. 2e) is found to be effective as a cytotoxic agent against the human acute lymphoblastic cell line that is CCRF-CEM. Eucalypglobulusal A (Fig. 2f), having a distinctive molecule characterised by a phloroglucinol nucleus coupled with a rearranged sesquiterpene skeleton, is reported for inhibitory activity on DNA topoisomerase 1(Top-1), where Top-1 catalysis gets inhibited, therefore it delays Top-1 position-mediated DNA double-strand damage[33].

Eucalyptal D (Fig. 2 g) has a synergistic effect in anticandidal activity in treating fluconazole-resistant *Candida albicans*[35]. Eucarobustol E (Fig. 2 h) is reported for its antifungal activity on *Candida albicans*. It acts by blocking the yeast to hypha transition & reducing the cellular surface hydrophobicity of the biofilm cells [34].



(a)



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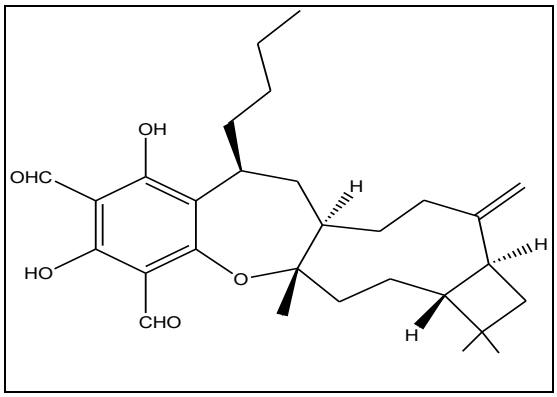

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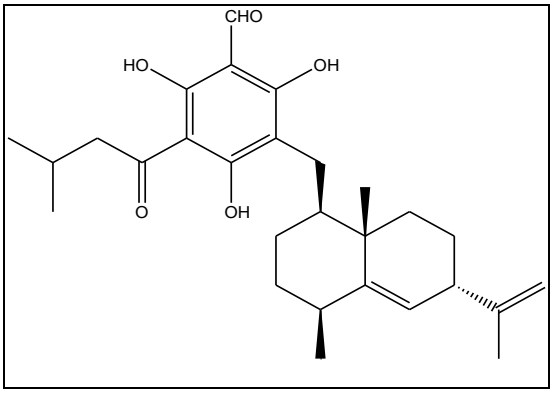
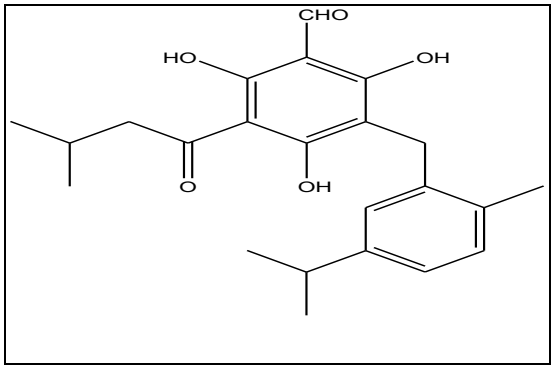
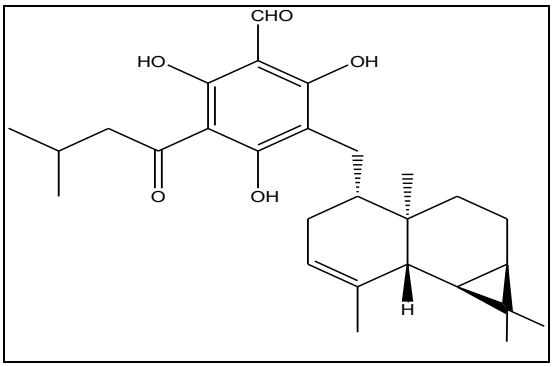


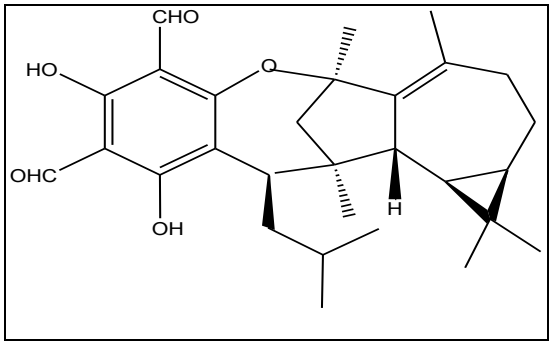
(d)



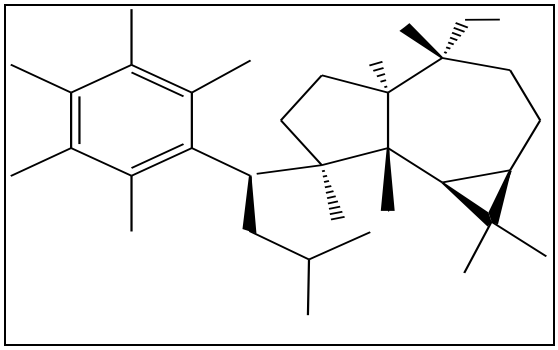
(e)



(f)



(g)



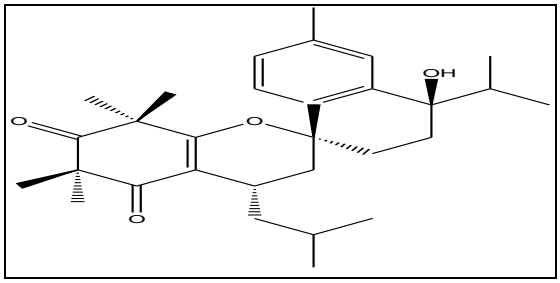
(h)

Fig 2. Formyl-Phloroglucinol–Terpene Meroterpenoids (a) Littordials B (b) Littordials C (c) Littordials E (d) Eucalrobusone C (e) Eucalypglobulusal F (f) Eucalypglobulusal A (g) Eucalyptal D (h) Eucarobustol E

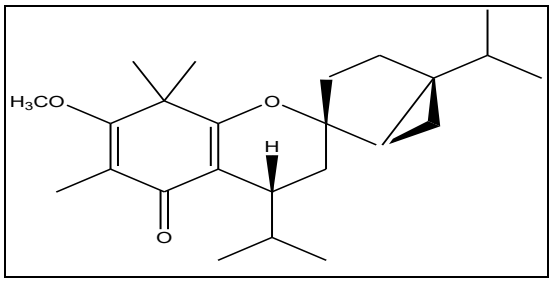
4.1.2 Polymethylated Phloroglucinol

Baeckfrutones A-S and rhotomentodiones C-E (Fig. 3a) are the most abounded polymethylated phloroglucinol in nature and are isolated from the aerial parts of *Baeckea frutescens* and *Rhodomyrtus tomentosa,* respectively. Baeckfrutone A contains rare enone-type phloroglucinol and a phellandrene unit, whereas baeckfrutones B-D, F, and I (Fig. 3 b-d) are the hybrids of enone-type phloroglucinol and sabinene moieties with different stereochemistry. Moreover, baeckfrutone M contains a 6/6/5/3 tetracyclic skeleton attachment with phloroglucinol moiety, and baeckfrutones O-P are unique hydroxytasmanone-phloroglucinol meroterpene hybrids [36][37][38]. Among these compounds, (-)-baeckfrutones B, J (Fig 3 e), and K (Fig 3 f) are reported for excellent cytotoxicity against DU145 and HCT116 cells while baeckfrutones F, N (Fig 3 g), G (Fig 3 h), (+)-I, J and S (Fig 3 i) are reported for their potent anti-inflammatory activity [36-37]. Among rhotomentodiones C-E, rhotomentodione D showed AChE inhibitory and antibacterial activity against Propionibacterium acnes[38].

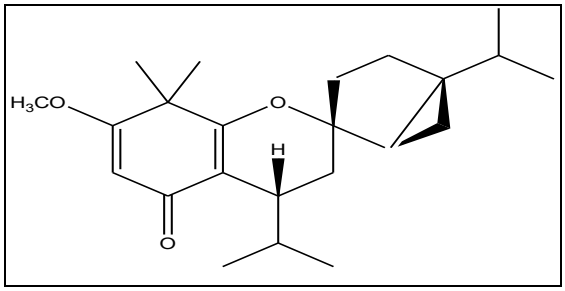
Enantiomers (±)-hyperjaponols H, two pairs of epimers hyperjaponols J-M, as well as hyperjaponol I, six new hetero-Diels-Alder adducts of polymethylated phloroglucinol and sesquiterpene giving shape to filicinic acid-based meroterpenoids, along with 6/6/10 ring skeletons, are reported from the *Hypericum japonicum*[39].


OH
O
O
O

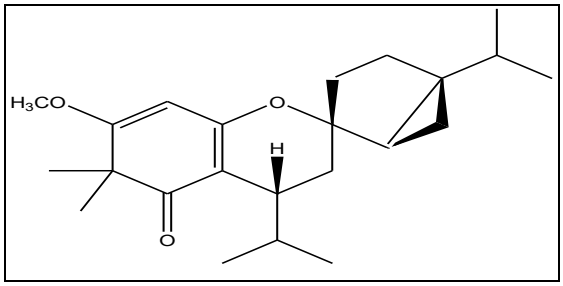
(a)


H3CO
O
H
O

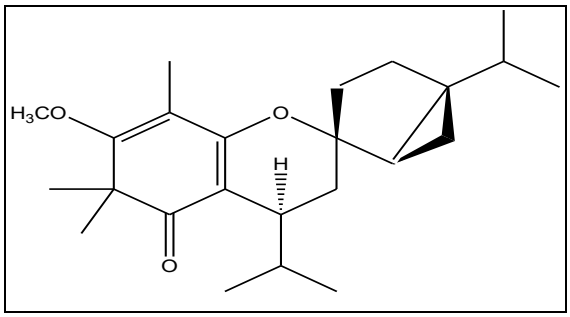
(b)


H3CO
O
H
O

(c)


H3CO
O
H
O

(d)


H3CO
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H
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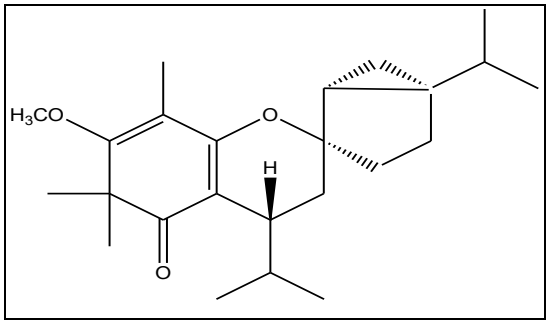
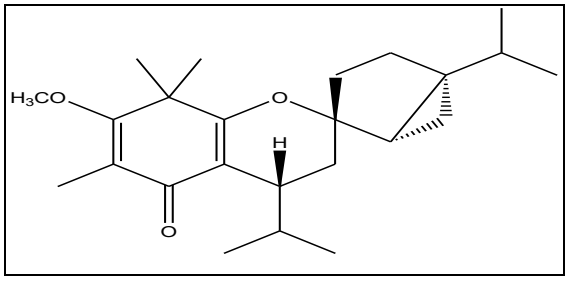
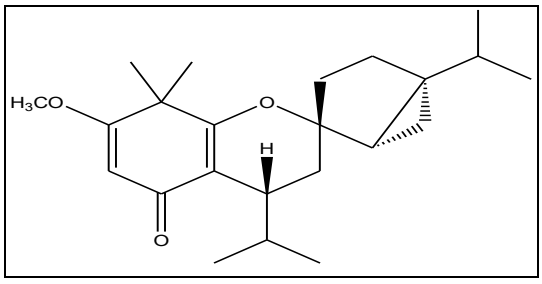
(e)


H3CO
O
H
O

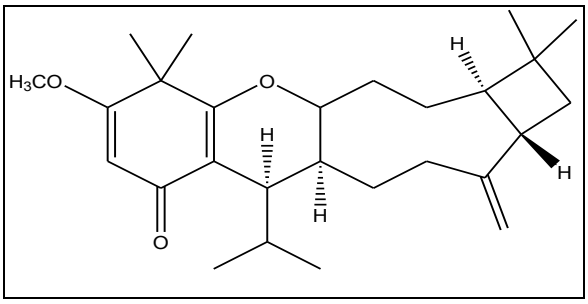
(f)


H3CO
O
H
O

(g)


H3CO
O
H
O

(h)

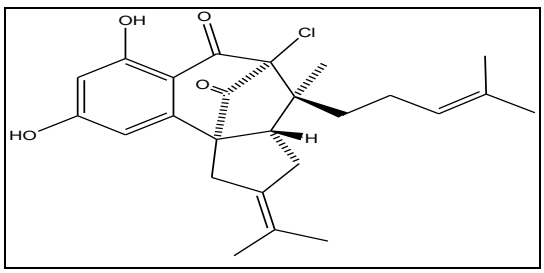


(i)

Fig 3. Polymethylated Phloroglucinol (a) Rhotomentodiones D (b) Baeckfrutone B (c) Baeckfrutone F (d) Baeckfrutone I (e) Baeckfrutone J (f) Baeckfrutone K (g) Baeckfrutone N (h) Baeckfrutone G (i) Baeckfrutone S

4.1.3 Merochlorins

Merochlorins are chlorinated meroterpenoids and a rare class of meroterpenes in terms of structure. Merochlorines A-F (Fig. 4 a-e) and meroindenon are identified as merchlorins to date from various marine strains of *Streptomyces sp*. Merochlorine A has four stereocenters surrounded by bicycle [3.2.1] octanone and chlorine attachment at the C-2 position, merochlorine B possesses a chlorinated vinylogous ester and an equally intimidating tetracyclic ring system with three contiguous stereocenters, and both compounds have novel terpene side chains. Merochlorine C has an uncommon macrocyclic ether structure[40][41][42]. Merochlorines E-F and meroindenon have a tetrahydroxynapthalene centre and a C-15 isoprene unit, along with a unique spiro γ-lactam functional group, and assemble a distinctive isobutyryl polyketide derived from (E)-4-methyl-2-pentenoic acid[43]. Merochlorines A-B have strong action against the methicillin-resistant *S. aureus* strain, while merochlorine C showed weak antimicrobial property. Merochlorines E-F showed strong antibacterial activities against *B. subtilis*, *K. rhizophila,* and *S. aureus*[43]. Meroindenon (Fig. 4f) exhibits weak antibiotic activity against the above-mentioned gram-positive bacteria[42][44].



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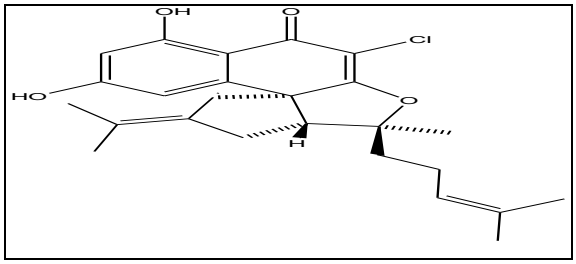

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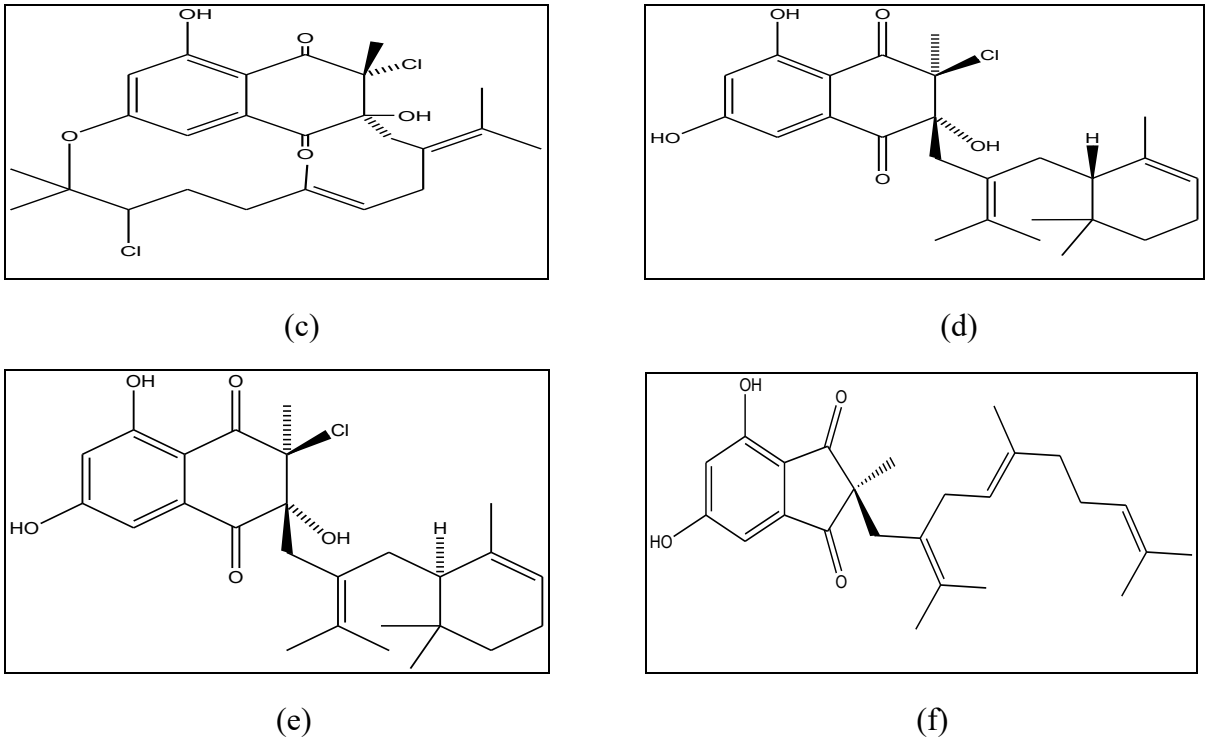


Fig 4. Merochlorins (a) Merochlorines A (b) Merochlorines B (c) Merochlorines C (d) Merochlorines E (e) Merochlorines F (f)Meroindenon

4.1.4 Naphthoquinone-Based Meroterpenoid

Naphthoquinone-based meroterpenoids contain 1,3,6,8-tetrahydroxynaphthlene (THN) core formed by a type III polyketide synthase (PKS) using malonyl-CoA as a substrate[45]. Tetrahydroxynaphthalene (THN) derived meroterpenoids can be divided into two classes, that is, class I and class II, Class I has C2 or C4 prenylation, and class II has C3 prenylation. Most of the naphthoquinone-based terpenoids belong to the class II[46]. Naphthoquinone-based meroterpenoids is a relatively common secondary metabolite produced by actinomycetes[47].

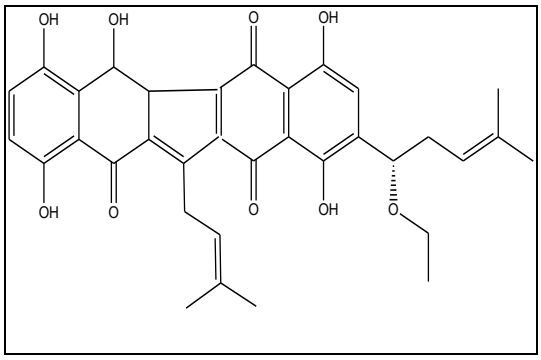
Shikometabolin H (Fig. 5a) is a dimeric naphthoquinone-based meroterpenoid isolated from the root of *Arnebia euchroma.* Shikometabolin H is reported for its significant anti-proliferative activities in human melanoma cell lines A375 and A2053 and human colorectal adenocarcinoma cells HCT116 and SW620[48]. Naphterpins and marinones have a rare aromatic oxidation pattern which is biosynthesised from THN[49].

Flaviogeranin B1 and B both have an amino group, which is connected at the C8 site of THN, while flaviogeranin D has an intact C-geranylgeranyl residue connected at C2 of THN, and the O-geranylgeranyl group of flaviogeranin B is connected with the hydroxyl on the C2 site of THN[45]. Flaviogeranin C2 (Fig. 5b) and flaviogeranin D (Fig. 5c) both are reported for potent antimicrobial activity on *Staphylococcus aureus* and *Mycobacterium smegmatis*; similarly, both compounds are reported for their cytotoxic activity on HeLa and A549 cell lines [45]. *Streptomyces sps*. CNQ-509 has potential for biosynthesis of hybrid isoprenoids, which further produce naphterpin, nitropyrrolin, and marinophenazine[50].

Naphthablins A-C, weak cytotoxic naphthoquinone meroterpenoids, are isolated from marine *Streptomyces* sp. CP26-58[47]. Prenylated naphthoquinone, 5, 8-O-dimethyl-11-deoxyalkannin (Fig. 5d) from *Arnebia euchroma* possesses a substantial cytotoxic activity on human cancer cell lines MG-63 and SNU387 [51].

Cordiaquinones A-B (Fig. 5 e-f), J-K (Fig. 5 g-h) obtained from the roots of *Cordia curassavica* are active against *Cladosporium cucumerinum* and *Candida albicans* for both the fungi and against larva of the yellow fever-transmitting mosquito *Aedes aegypti*[52]. Cordiaquinone L and M, obtained from the roots of *Cordia leucocephala,* are structurally similar, where cordiaquinone L is 6-[10-(12,12-dimethyl13β-cyclohexyl) ethyl]-1,4-naphthalenedione and cordiaquinone M is 5-methyl-6-[10-(12,12-dimethyl13β-hydroxy-16-methenyl-cyclohexyl) methyl]-1,4-naphthalenedione[53].

Microphyllaquinone is a well-known naphthoquinone meroterpenoid derived from the root of *Cordia globose*[54].



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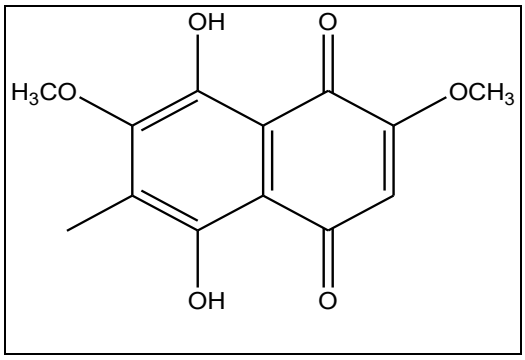

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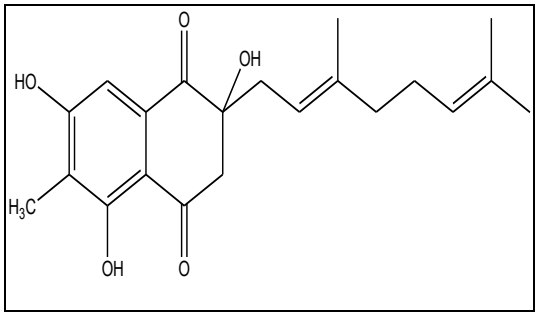

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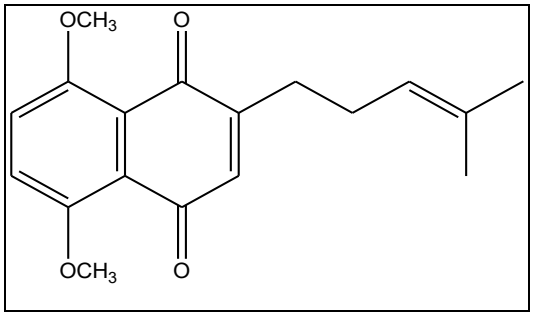

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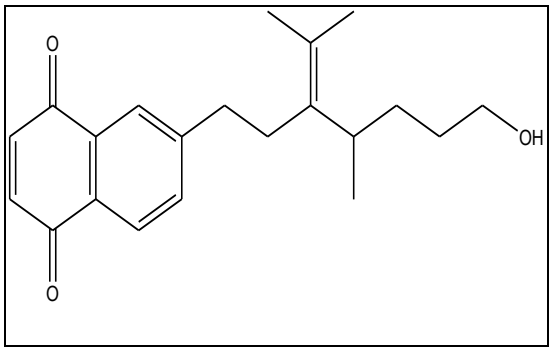

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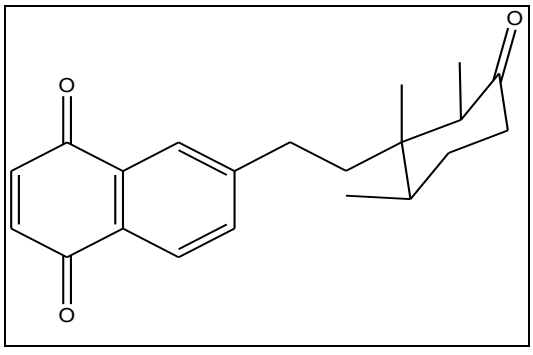

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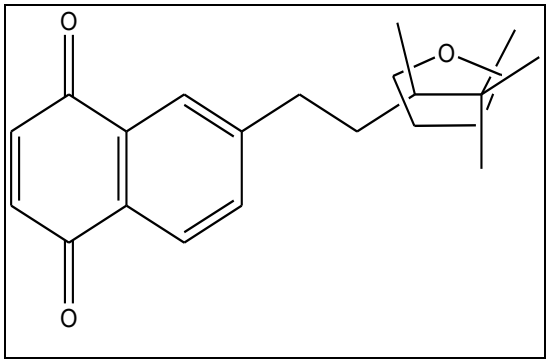

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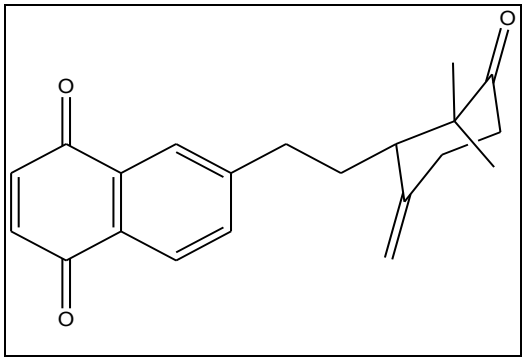


(h)

Fig 5: Naphthoquinone-Based Meroterpenoid (a) Shikometabolin H (b) Flaviogeranin C2 (c) Flaviogeranin D (d) 5, 8-O-dimethyl-11-deoxyalkannin (e) Cordiaquinones A (f) Cordiaquinones B (g) Cordiaquinones J (h) Cordiaquinones K

4.2 Plant-Derived Meroterpenoid

Meroterpenoids with a resorcinyl core specifically substituted with isoprenyl, alkyl, or aralkyl side chains at the para position are known as phytocanabinoids or cannabinoids. In the chemistry of cannabinoids, the alkyl side chain particularly has an odd number of carbons, such as orcinoids having one carbon, varinoids having three, and olivetoids having five carbons[55].

Major phytocannabinoids are obtained from *Cannabis sativa L.* (Cannabaceae). It extensively produces alkyl-type cannabinoids which has an isoprenyl moiety and a pentyl side chain. Cannabigerols, cannabichromenes, cannabidiols, (-)-Δ9-trans-tetrahydrocannabinols (Fig. 6a), (-)-Δ8-trans-tetrahydrocannabinols, cannabiocyclos, cannabielsoins, cannabionols, cannabinodiols (Fig. 6 b), cannabitriols, etc., are a class of cannabinoids generally obtained from this plant[56][57].

Various species of *Rhododendron* (Ericaceae), *R. dauricum, R. adamsii, R. anthopogonoides* Maxim, and *R. rubiginosum* have been reported to have cannabichromene (Fig. 6c) (CBC) attached with an orcinol side chain that is a methyl group[58]. *R dauricum* has been reported to produce cannabinoids grifolic acid, daurichromenic acid, confluentin, and rhododaurichromenic acids A and B, having a sesquiterpene moiety[59]. Similarly, cannabigerol cynic acid, cannabigerorcynic acid methylester, daurichromenic acid (Fig. 6d), and chromene meroterpenoids are isolated from *R. adamsii*[60]. *R. anthopogonoides* is noted to contain cannabinoid-like chromane and chromene derivatives named anthopogocyclolic acid, anthopogochromenic acid, cannabiorcichromenic acid, and cannabiorcicyclolic acid[61][62]. Cannabiorcichromenic acid, decarboxycannabiorcichromenic acid, and rel- (6aS,10aR)-decarboxy- Δ9 -tetrahydrocannabiorcolic acid B are reported for dose-dependent growth restoring activity in the mutant yeast [64]. Anthopogochromenes A and B and rubiginosins A-G have been reported from *R. rubiginosum*[58].

Amorfrutins attached with cannabinoid skeleton having an aralkyl side chain, hence known as prenylated bibenzyls, have been reported from the flowering tops of *Helichrysum*

umbraculigerum Less. (Compositae), roots of *Glycyrrhiza foetida* Desf. and *Amorpha fruticose* L. (Fabaceae)[55][63][64].

Liverworts, generally known as hepatics, are a major source of cannabinoid compounds with a bibenzyl backbone, and the known compounds are lunularic acid and vittatin[65]. Cannabinoids with a similar backbone have also been reported in plants belonging to the Radulaceae family, such as *Rudula perrotteii Gottsche ex* Steph[66], *Rudula marginata* Taylor ex Gottsche[67][68], and *Rudula laxiramea* Steph.

Along with plants, cannabinoids are also found in a few fungal organisms, such as mycorrhizal fungi belonging to the genus Albatrellus and family Albatrellaceae, and another species is *Cylindrocarpon olidum* belonging to the Nectriaceae family. Mycorrhizal fungi produce grifolic acid (GFA) and its derivatives like grifolin, neogrifolin, and confluentin[69][70]. Canabiorcichromenic acid and a halogenated cannabinoid, 8-chlorocanabiorcichromenic acid, are noted to occur in *C. olidum*[71].

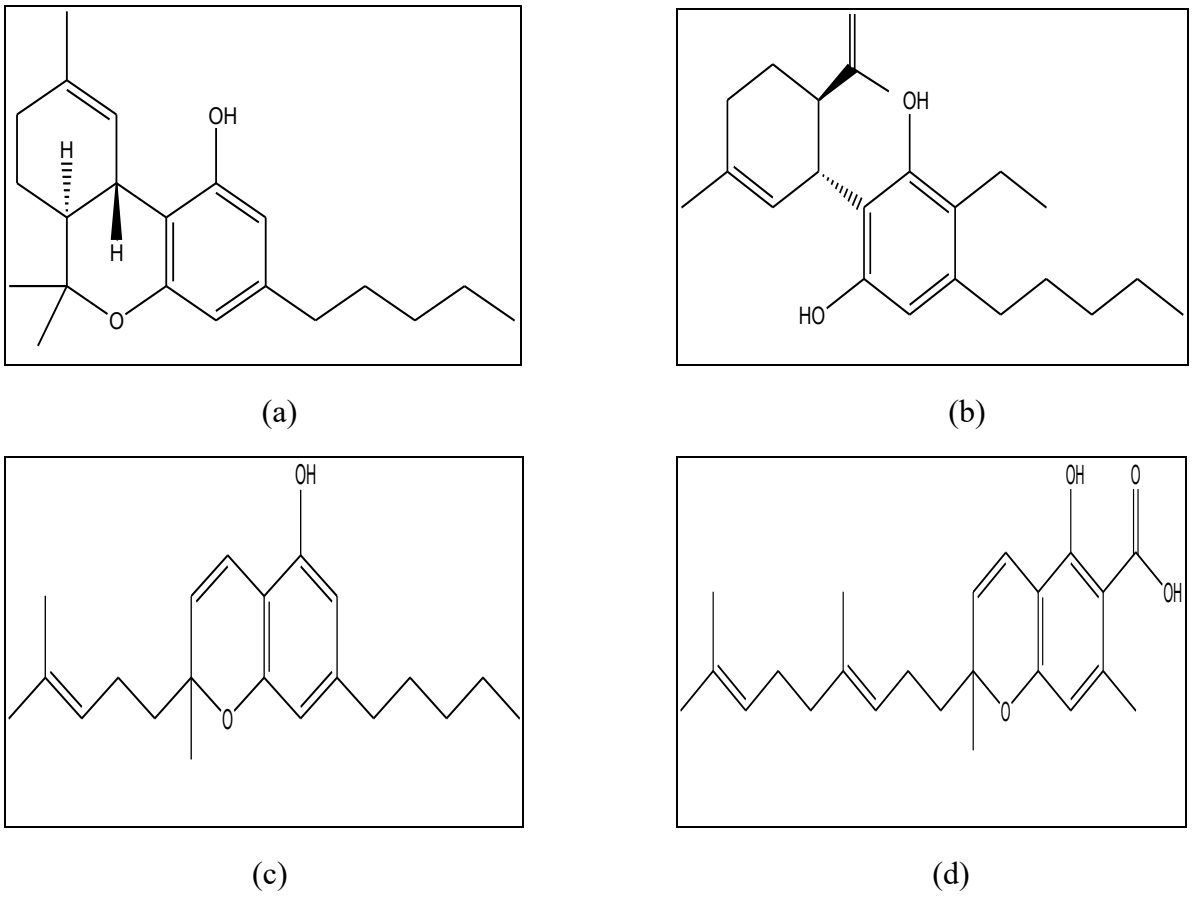


Fig 6: Plant-Derived Meroterpenoid Cannabinoids (a) Tetrahydrocannabinols (b) Cannabinodiols (c) Cannabichromene (d) Daurichromenic acid

4.2.1 Polycyclic Meroterpenoid

The Polycyclic meroterpenoids class usually contains congested polycyclic skeletons like berkeleyacetals with 6/7/6/5/6 cyclic system, 6/6/6/6 cyclic talarolutins, and many more. There are many polycyclic meroterpenoids isolated and evaluated for their activity[72]. Talarolutins

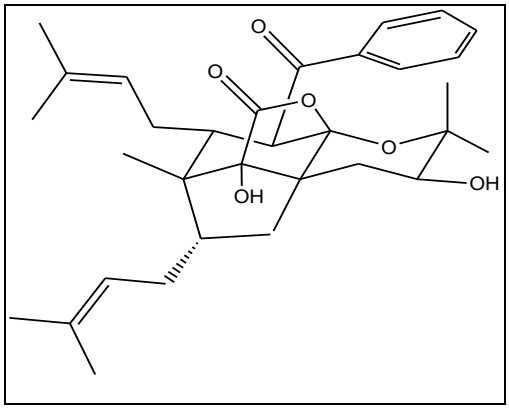
A–D have been characterised from an endophytic fungus isolated from Talaromyces minioluteus (G413), found on the leaves of *Silybum marianum* (L.)[73].

Hyperinoids A (Fig. 7a) and B (Fig. 7 b), having potent NF-kB pathway inhibitory activity, were isolated from the leaves & twigs of *Hypericum patulum* and are prenylated acylphloroglucinol type of meroterpenoids, where both of them contain respective two unique tetracyclic systems of 11,12-dioxatetracyclo [5.4.3.0.0] tetradecane and 10,11-dioxatetracyclo [5.3.3.0.0] tridecane[74].

Magterpenoids A-C (Fig. 7 c-e) exhibiting potent PTP1B inhibitory activity are obtained from the bark of *Magnolia officinalis var. biloba*, where magterpenoid A has a unique 4,6,11-trioxatricyclo [5.3.1.0] undecane skeleton with an uneven meroterpenoid moiety, magterpenoid B has an extraordinary 6/6/6/6 polycyclic backbone, & magterpenoid C has a novel terpenoid moiety substituted with quinone at the C6-C3 unit[75].

Ganoleucin D, a phenolic meroterpenoid with a C-15 side chain and spiroapplanatumine K (Fig. 7f) and L (Fig. 7 g), (±)-spirolingzhine A and spirolingzhine D compounds with spiro rings were isolated from the fruits of *Ganoderma leucocontextum*[76]. (±)-cochlearins A-E, F-I showed antioxidant activity [76]. Ganoleucin D, spiroapplanatumine L, and spirolingzhine D exhibit aldose reductase inhibitory activity in bovine lens tissue. Also, Ganoleucin D, spiroapplanatumine K and L, (±)-spirolingzhine A and spirolingzhine D exhibited HMG-CoA reductase inhibitory activity and anti-cancerous activity against HCT-116 and K562 cell lines[76]. Polycyclic meroterpenoids ganotheaecoloids A-N and petchiene A were tested for COX-2 inhibitory activity. Ganotheaecoloids J exhibited potent inhibitory activity[4]. (±)-Ganoapplanin isolated from *Ganoderma applanatum* contains an unprecendented dioxaspirocyclic skeleton built from a 6/6/6/6 tetracyclic system & a rare tricycle [4.3.3.0] dodecane. Based on structure it contains polycyclic ring structure and 2,5-dihydroxybenzoyl fragment[77]. Enantiomers of Applanatumines B-D from *Ganoderma applanatum* are active JAK3 inhibitors. Also, Applanatumine B is active towards discoidin domain receptor 1(DDR1)[78].

Ganocins A-D (Fig. 7 h) and (±)-Cochlearols A & B (Fig. 7i) are obtained from the fruits of *Ganoderma cochlear*. Cochlearol A contains a rare dioxaspirodecane moiety, while ganocin A-C have a spirodecane ring system, and ganocin D has an eight-membered ring[79]. (-)-Cochlearol B possesses strong antifibrotic activity in a dose-dependent way & also strongly inhibits pSmad and shows protective effects against TGF-β1–induced injury in rat renal proximal tubular cells[79] while Ganocin D reported for its weak anti-AChE effect[80]. (±) Gancochlearols A & B, 2 pairs of cytotoxic dimeric mertoterpenoid enantiomers isolated from the fruiting bodies of *Ganoderma cochlear,* are reported to exhibit cytotoxic effect on A579, K562, & Huh-7 human cancer cell lines and inhibit COX-2 expression[3].



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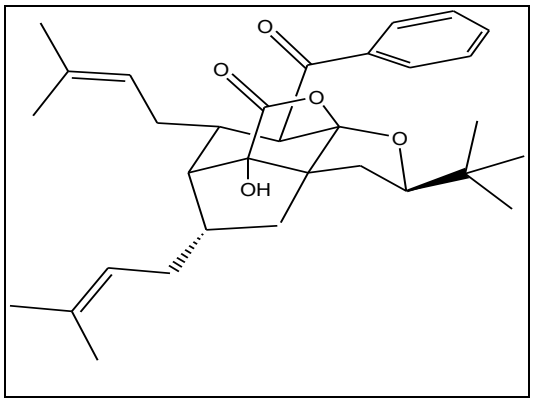

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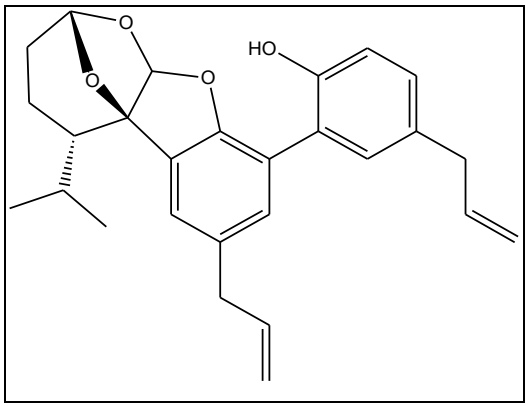

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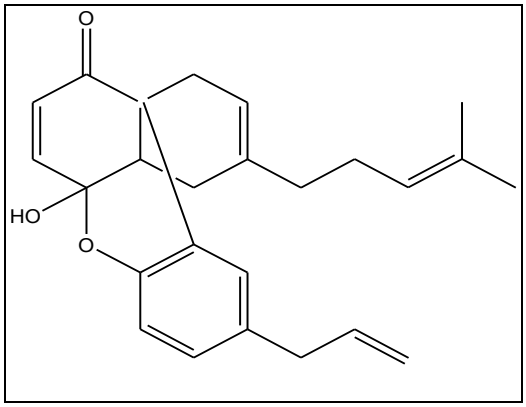

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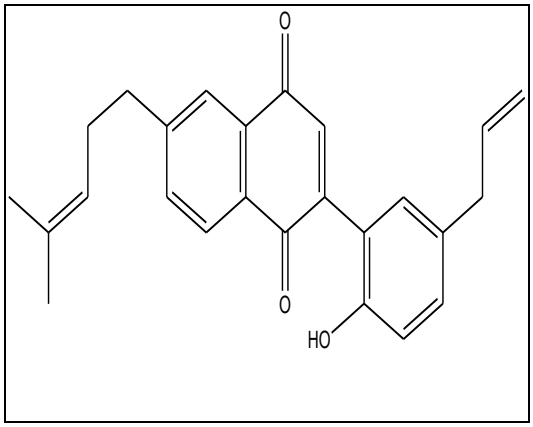

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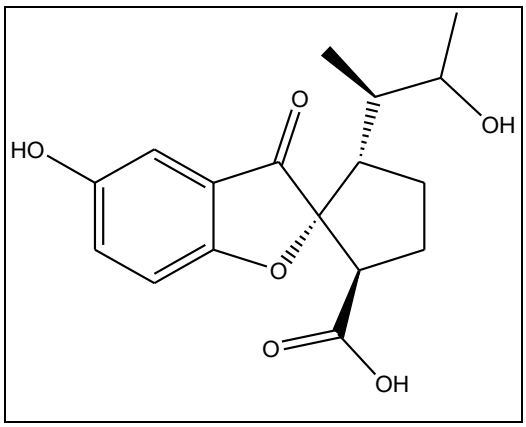

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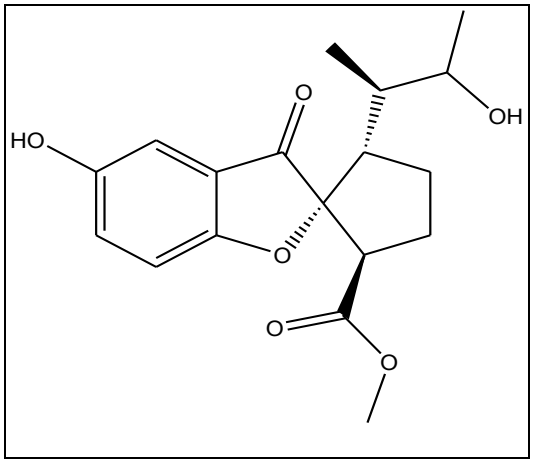

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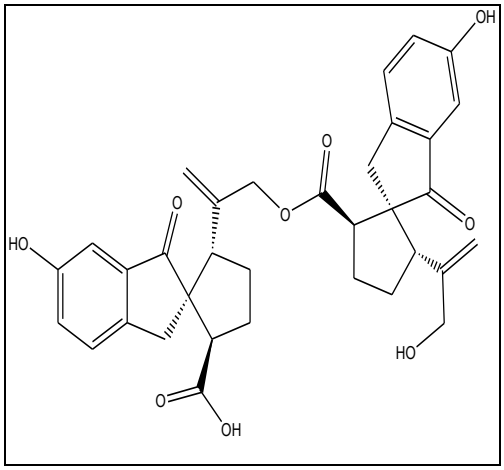

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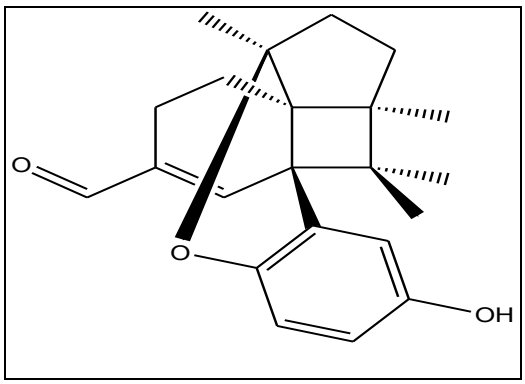


(i)

Fig 7: Polycyclic Meroterpenoid (a) Hyperinoids A (b) Hyperinoids B (c) Magterpenoids A (d) Magterpenoids B (e) Magterpenoids C (f) Spiroapplanatumine K (g) Spiroapplanatumine L (h) Ganocins D (i) Cochlearols B

4.2.2 Tropolone Sesquiterpenoids

Tropolone sesquiterpenoids consist of a main 11-membered macrocycle, which is obtained from humulene and attached to one or two dihydropyran rings that act as a bridge between the macrocycle and polyketide-derived tropolones. Advanced modification can be done by hydroxylation, various olefin configurations of the central humulene macrocycle, replacement of tropolone moieties via monobenzopyranyl, and oxidative ring contraction of the polyketide[81].

Pycnidione, xenovulene A (Fig. 8a), and eupenifeldin are the main reported tropolone sesquiterpenoids obtained from *Theissenia rogersii* 92031201 (81), *Acremonium strictum* IMI 501407[82], and *Eupenicillium brefeldianum*[83] respectively. Other examples with unknown sources are neosetophome B (Fig. 8b), xenovulene A, and epolone A (Fig. 8c)[81] Pycnidione (Fig. 8d) is reported for its antiproliferative activity on human lung cancer cells, xenovulene A is reported for the inhibition of the human γ-aminobutyrate A (benzodiazepine) receptor, eupenifeldin (Fig. 8e) and neosetophome B are reported for antitumor activity at nanomolar

concentration, and epolone A is reported for selective induction of erythropoietin expression at a micromolar concentration in human cancer cell lines[81].

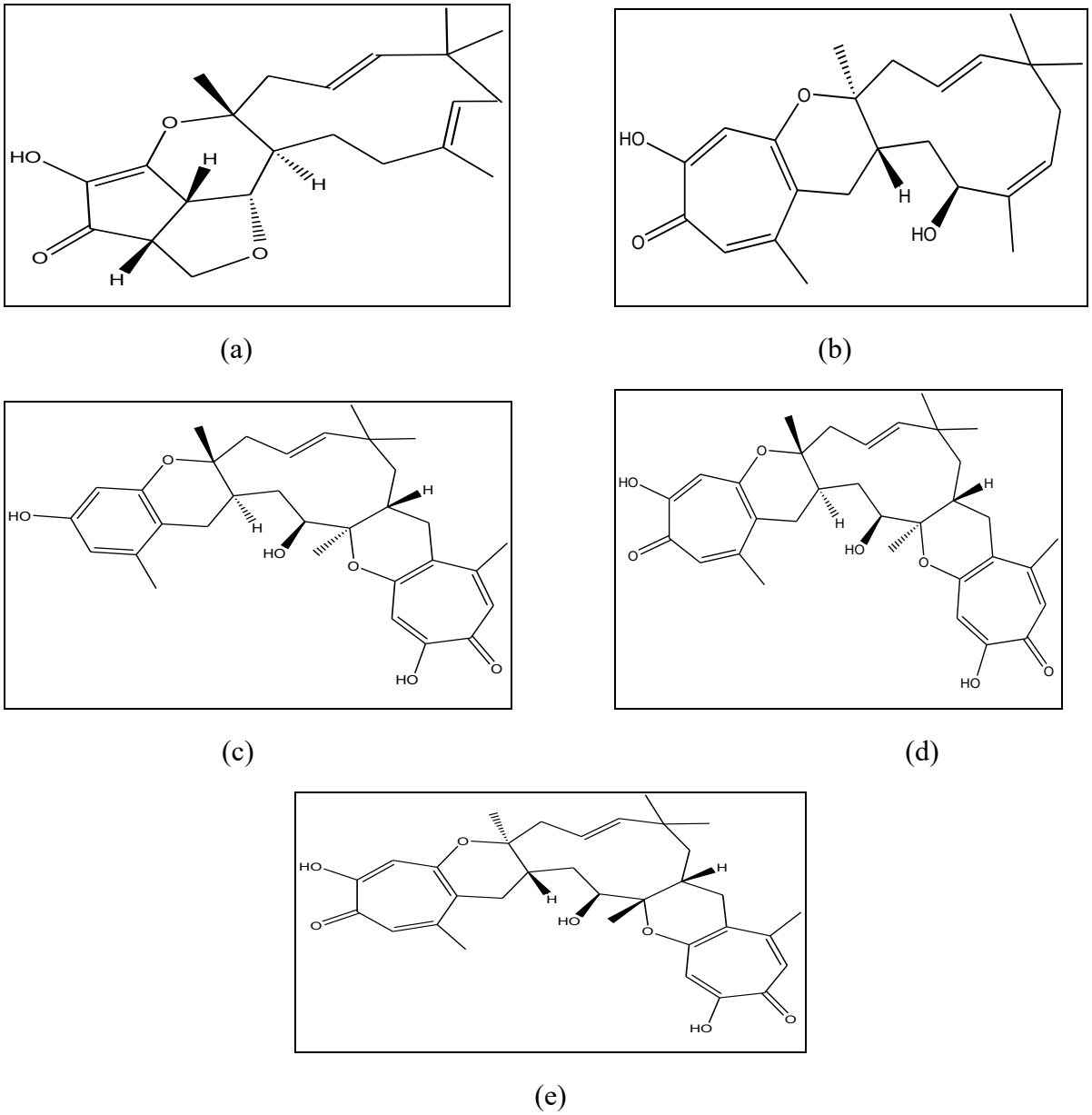


Fig 8: Tropolone Sesquiterpenoids (a) Xenovulene A (b) Neosetophome B (c) Epolone A (d) Pycnidione (e) Eupenifeldin

4.3 Marine Fungal Meroterpenoids

Acyclic merohemiterpenoids such as Mycophenolic acid–based merohemiterpenes (7–17) isolated from Penicillium bialowiezense, a coral-associated fungus from Sarcophyton subviride, were found to inhibit inosine-5′-monophosphate dehydrogenase (IMPDH2) and to exert in vitro immunosuppressive effects on T-lymphocyte proliferation[84].

Out of the three compounds, acremine S, acremine T & lumichrome, lumichrome are cyclic merohemiterpenes obtained from the cultures of the marine fungi *Acremonium persicinum* and the marine sponge *Mycale* sp. Lumichrome showed strong inhibition of acetylcholinesterase

(AChE) near to standard used (85). Whereas acremine T inhibited butyrylcholine esterase (BuChE) near to standard galantamine[85].

Guignardone A, 12-hydroxylated guignardone A, guignardone J, and guignardone M are meromonoterpenoids that were reported for their antimicrobial activity on *P. aeruginosa, E. faecalis* & methicillin-resistant *S. aureus*. Only Guignardone A shows activity on *P. aeruginosa* and methicillin-resistant *S. aureus*[14].

Alternarin A, a rearranged drimane meroterpenoid characterised by a thioglycerate moiety isolated from the coral-linked fungi Alternaria species ZH-15, which shows concentration-dependent inhibition of spontaneous Ca^{2+} oscillations (SCOs) via decreasing both SCO frequency and amplitude. In addition to its Alternarin A effectively suppressed seizureogenic agent-induced hyperactive SCOs frequency and amplitude, respectively, in cortical neurons[86].

7-Deacetoxyyanuthone A (Fig. 9a), 2,3-hydrodeacetoxyyanuthone A (Fig. 9 b), and 22-deacetylyanuthone A (Fig. 9c) are acyclic merosesquiterpenoids isolated from the marine fungi Gliomastix species ZSDS1-F7, obtained from the marine sponge *Phakellia fusca*. Thiele exhibited *in vitro* cytotoxicity against various cell lines of breast cancer, prostate cancer, myeloid leukaemia, non-small lung cancer, gastric cancer, lung carcinoma, and acute lymphoblastic leukaemia. 7-Deacetoxyyanuthone A possessed antitubercular activity[87].

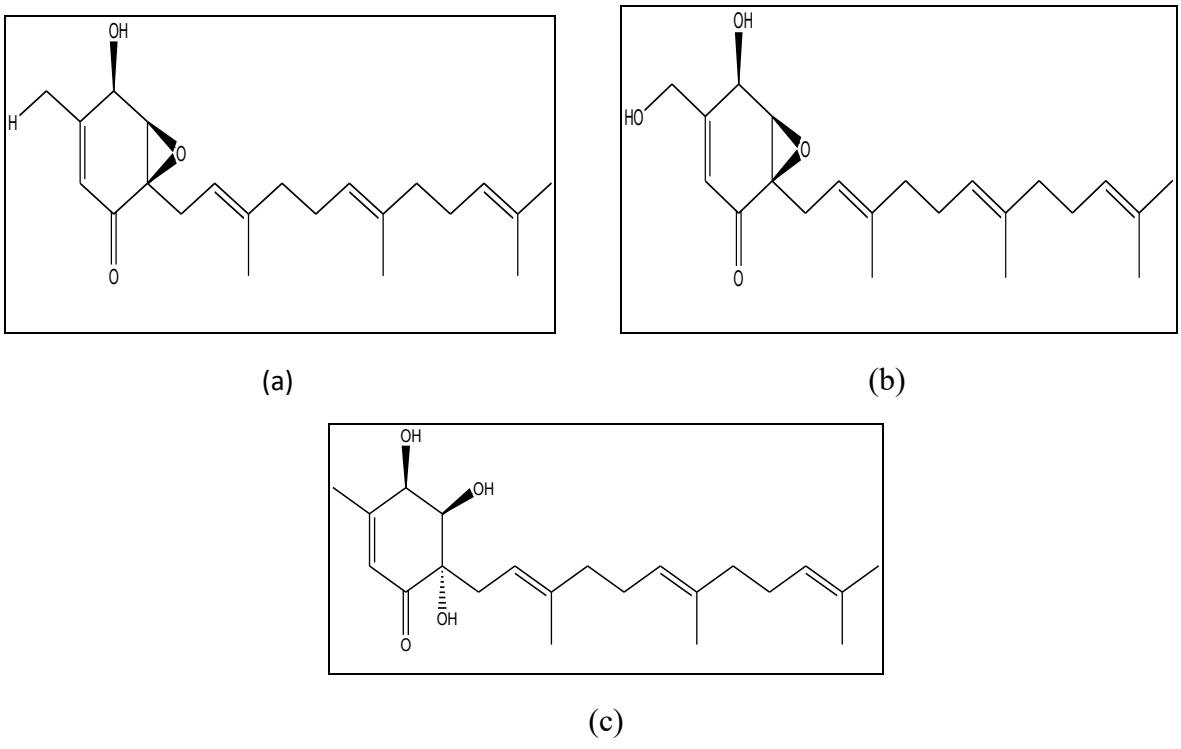
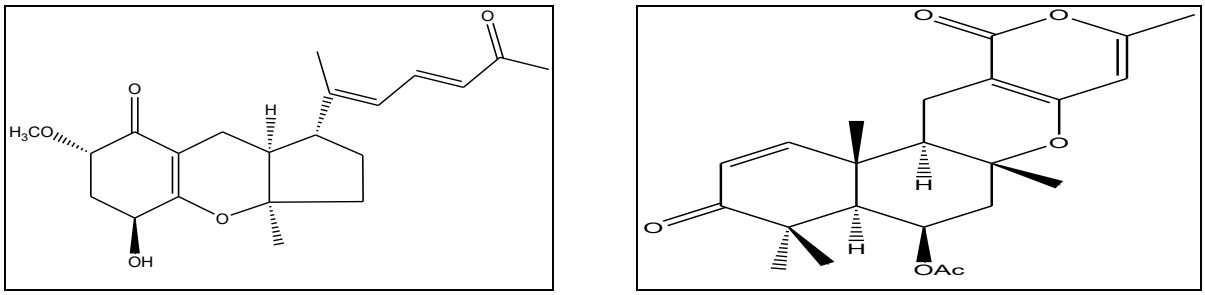


Fig 9: (a) 7-Deacetoxyyanuthone A (b) 2,3-hydrodeacetoxyyanuthone A (c) 22-deacetylyanuthone A

Chrysomutanin with a chain sesquiterpene unit derived from the *Penicillium chrysogenum* LD-201810 shows a strong cytotoxic effect on the HL-60 cell line[10]. Monocyclic merosesquiterpenoid guignardone L (Fig. 10a) is also obtained from the mangrove endophytic fungi *A. flavipes*, which doesn't show significant antibacterial or cytotoxic activities[88].

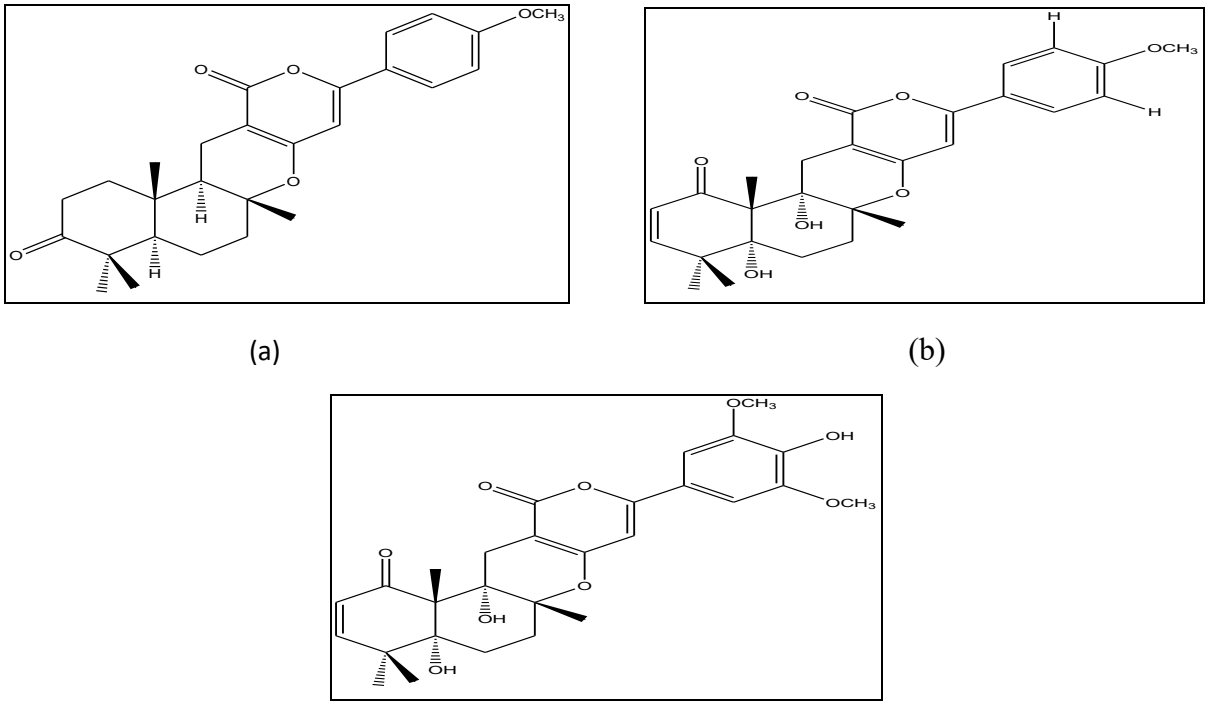
Bicyclic (drimane) merosesquiterpenoid linked to a 2-pyrone includes asperversins A-G (Fig. 10 b) featuring a 5/6/6/6 ring system, along with their analogues asperversins C–G, which possess a 7/6/6/6 cyclic ring framework, derived from the marine fungus *Aspergillus versicolor* isolated from South China Sea sediment, are seen to exert AChE inhibitory activity, asperversin G[89][90][91].



(a) (b)

Fig 10: (a) Guignardone L (b) Asperversins G

3-Epi-arigsugacin E (Fig. 11a), arisugacin B (Fig. 11 b), and territrem C (Fig. 11c), drimane merosesquiterpenoids which are linked to a phenyl 2-pyrone ring system, have been obtained from the mangrove-derived fungi Penicillium species, obtained from the leaves of the mangrove plant *Kandelia candel*. All 3 exhibit potent AChE inhibitory activity[93].



(a) (b)

(c)

Fig 11: (a) 3-Epi-arigsugacin E (b) Arisugacin B (c) Territrem C

Pyripyropenes like 11-deacetylpyripyropene O (Fig 12 a), pyripyropenes A (Fig 12 b) and O (Fig 12 c) isolated from the marine-derived fungus *Fusarium lateritium* 2016F18-1, which was isolated from the marine-derived fungus *Fusarium lateritium* 2016F18-1, which in turn was

obtained from the marine sponge *Phyllospongia foliascens*, as well as from *Neosartorya pseudofischeri* which were isolated from the inner tissue of the starfish *Acanthaster planc* are reported to possess prominent cytotoxicity against human cancer cell lines, for nasopharyngeal carcinoma (CNE1, CNE2 & HONE1), lung carcinoma (GLC82) and Hepatic carcinoma (HL7702)[94][95][96].

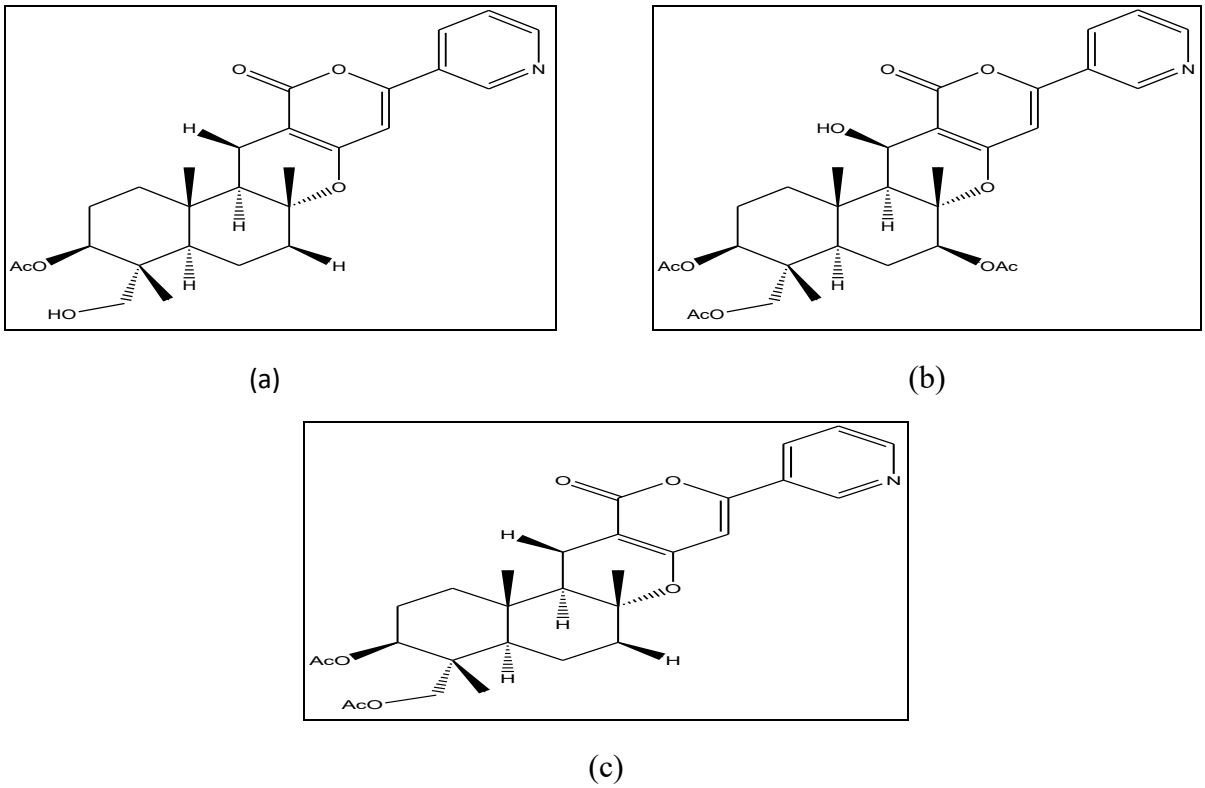


Fig 12: (a) 11-deacetylpyripyropene O (b) Pyripyropenes A (c) Pyripyropenes O

Epoxyphomalins A (Fig. 13a) and B (Fig. 13 b) are bicyclic merosesquiterpene linked to a 4-pyrone isolated from the culture extract of the marine-derived fungus *Paraconiothyrium cf. sporulosum*, obtained from the marine sponge *Ectyplasia perox* collected in the Caribbean Sea, and exhibiting significant antiproliferative activity against 36 human tumour cell lines[97].

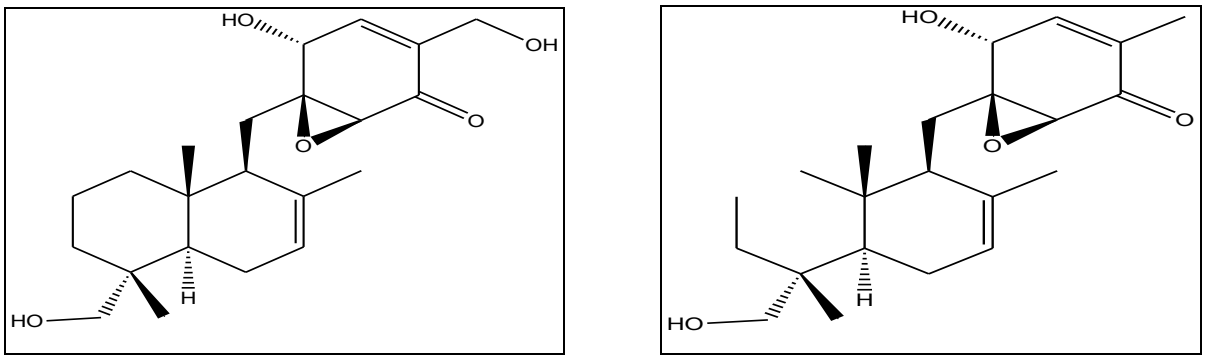
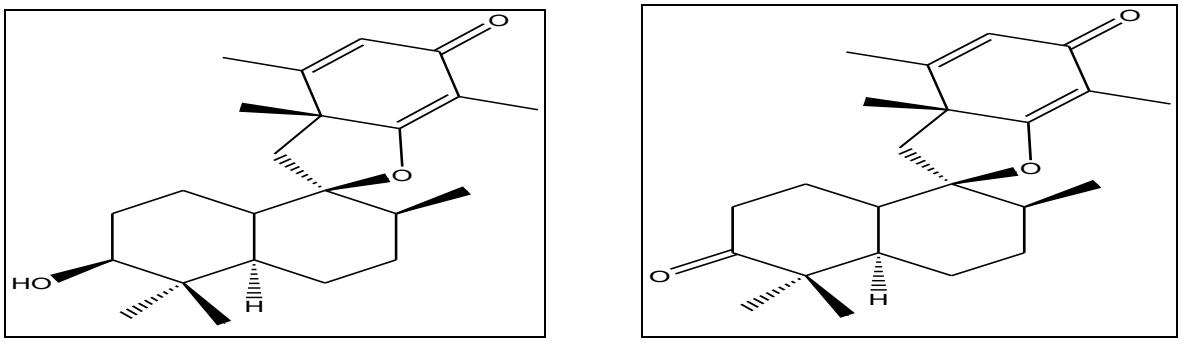


Fig 13: (a) Epoxyphomalins A (b) Epoxyphomalins B

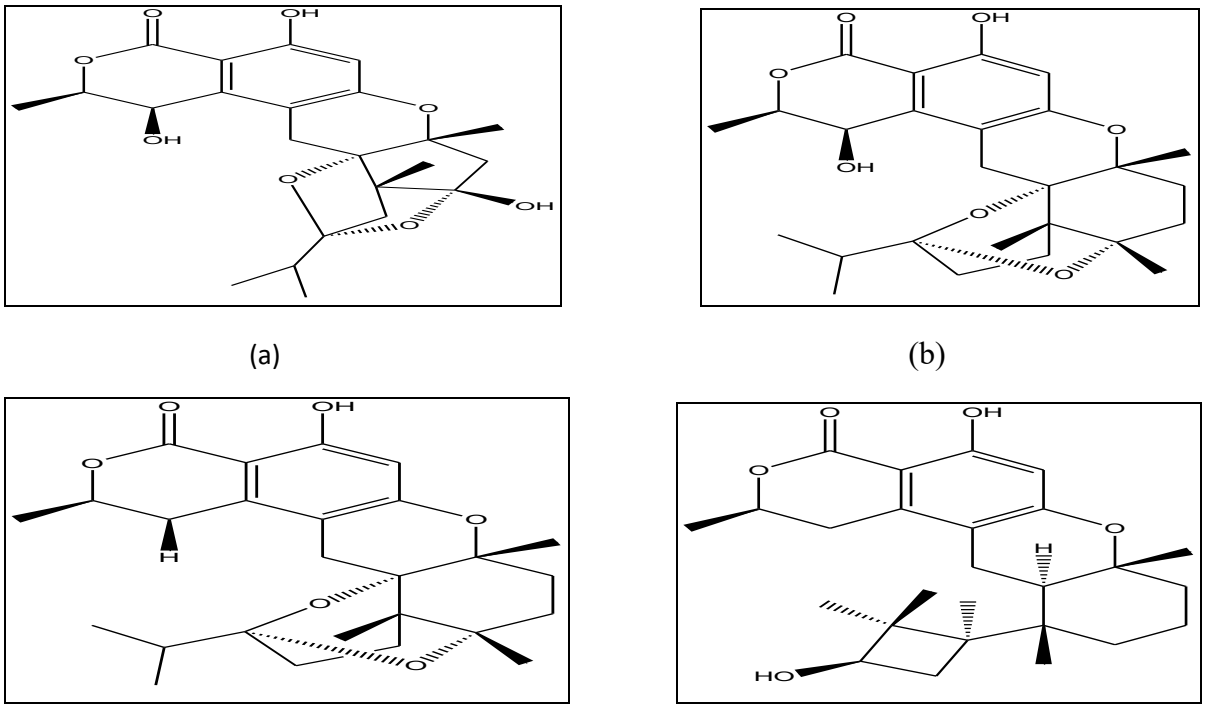
Drimane merosesquiterpeneoid linked to spirobezopyran derivative example chartarolides A–C (Fig. 14) isolated from the fermentation broth of the marine-derived fungus *Stachybotrys chartarum* WGC-25C-6, obtained from the marine sponge *Niphates recondita*, and exhibiting potent cytotoxicity against a panel of human tumor cell lines, as well as inhibitory activity toward four tumor-associated protein kinases. Chermesins A and B isolated from the culture extract of the algicolous fungus *Penicillium chermesinum* EN-480, obtained from the marine red alga *Pterocladiella tenuis*, and exhibiting activity against *Candida albicans*, *Escherichia coli*, *Micrococcus luteus*, and *Vibrio alginolyticus*[98][99].



(a) (b)

Fig 14: (a) Chartarolides A (b) Chartarolides B

Talaromyolides A-D (Fig 15) isolated from a marine fungus *Talaromyces sps.* CX11 are drimane merosesquiterpene linked to isochromone derivatives having a unique structure like berkeleyacetals with a 6/7/6/5/6 cyclic system, 6/6/6/6 cyclic talarolutins[100][72]. Talaromyolide D value shows antiviral activity against *Pseudorabies virus*.(72) Chrodrimanins O, R, and S show tyrosine phosphatase 1B (PTP1B) inhibitory activity[101].

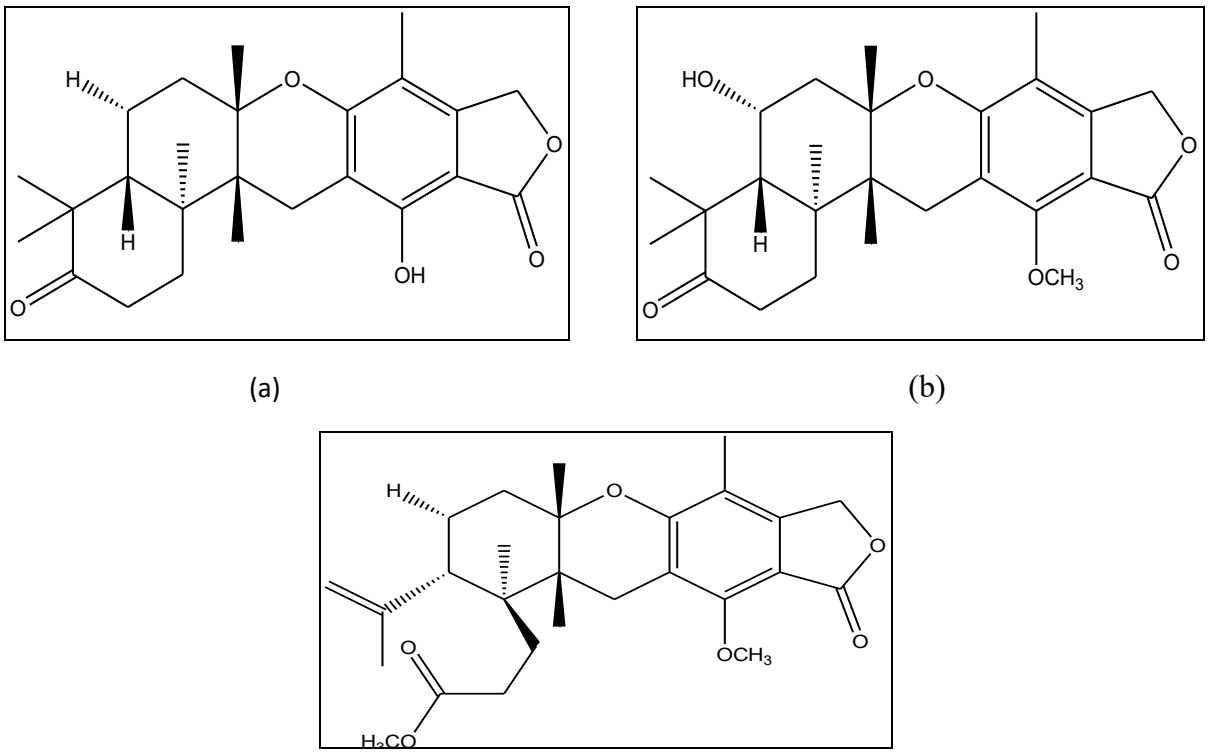


(a) (b)

(c) (d)

Fig 15: (a) Talaromyolides A (b) Talaromyolides B (c) Talaromyolides C (d) Talaromyolides D

Austalides V-X (Figure 16), drimane sesquiterpenes linked to 5-methylorsellinic acid obtained from a marine sediment derived fungi *Penicillium rudallense,* which shows potent inhibitory action on RANKL-induced osteoclast differentiation. Among all these compounds, Austalide V showed minor cytotoxicity[102].



(a) (b)

(c)

Fig 16: (a) Austalides V (b) Austalides W (c) Austalides X

Photeroids A and B, the first phenolic merosesquiterpenoid featuring a highly fused 6/6/6/6 tetracyclic ring system derived from the deep-sea derived fungi *Phomopsis tersa,* exhibited moderate cytotoxic activities[103].

Austinoid derivatives penicianstinoids A and B (Fig. 17a- b), belonging to drimane merosesquiterpenes linked to 3,5-methyloresellinic acid, are isolated from the culture extract of Penicillium species TGM112, obtained from the medicinal mangrove *Bruguiera sexangula var. rhynchopetala*, collected in the South China Sea. Both compounds inhibit the growth of newly hatched larvae of *Helicoverpa armigera Hubner* and *Caenorhabditis elegans*(15). Acetoxydehydroaustin A (Fig. 17c) and Austin (Fig. 17d) are found to affect sodium channel activation and inactivation at 50 µM concentration, where Austin is more effective than Acetoxydehydroaustin A. Acetoxydehydroaustin A and Austin also exhibit activity on window current[104].

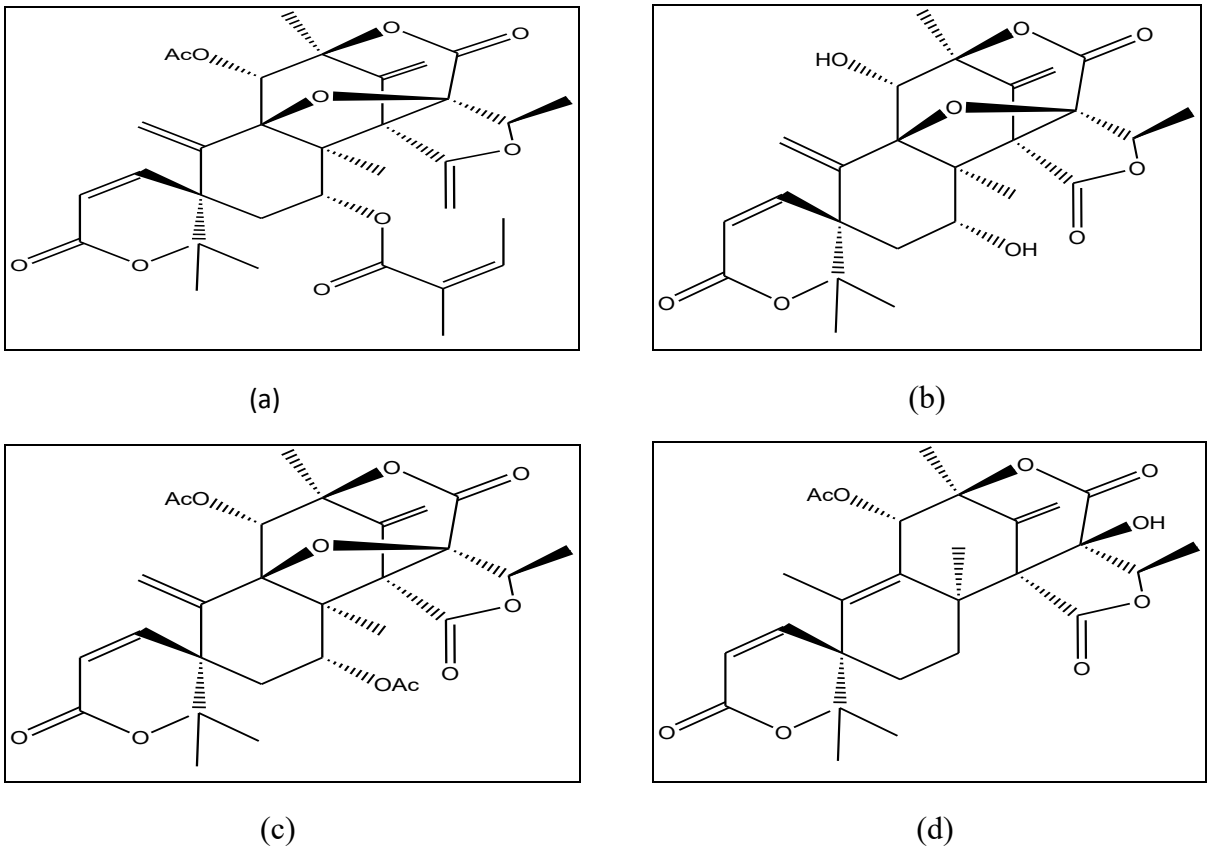


Fig 17: (a) Penicianstinoids A (b) Penicianstinoids B (c) Acetoxydehydroaustin A (d) Austin

Preaustinoids A6, a cyclic terpene ketone/ carbopolycyclic compound from *Penicillium sp.* SF-5497 is shown to have inhibitory activity on PTP1B in a non-competitive method & favourably binds to the free enzyme rather than to the enzyme substrate complex in a dose-dependent manner[105]. Berkeleyacetal A, 22-epoxyberkeleydione, 22-deoxyminiolutelide B, miniolutelide C, and berkeleydione were reported from *Penicillium* strain 303#. Compounds 22-deoxyminiolutelide B (Fig. 18a) and miniolutelide C (Fig. 18 b) exhibited moderate cytotoxic activity against the MDA-MB-435, HepG2, HCT-116, and A549 cell lines[106]. Preaustinoids 22-deoxyminiolutelide A, 4S-hydroxy-22-deoxyminiolutelide B, miniolutelide A, and miniolutelide B, isolated from the culture extract of the marine-derived fungus *Penicillium ubiquetum* MMS330 obtained from the blue mussel *Mytilus edulis*, exhibited cytotoxic activity against KB (keratin-forming tumour) and MCF-7 cell lines[105].

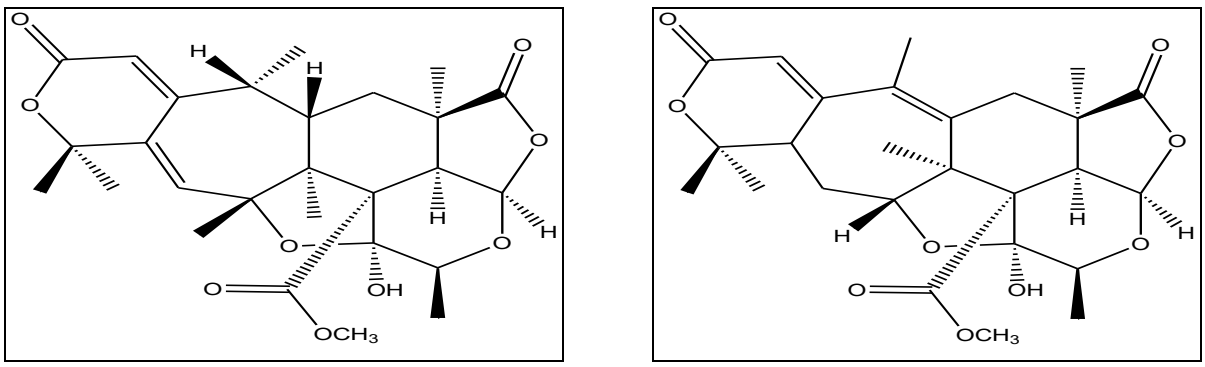


Fig 18: (a) 22-deoxyminiolutelide B (b) Miniolutelide C

Analogues of berkeleyacetals, amestolkolides A (Fig. 19) from the endophytic fungi *Talaromyces amestolkiae* YX1 were isolated from the fully grown leaves of the mangrove tree *Kandelia obovate*, and exhibit anti-inflammatory activity by inhibition of the LPS-activated NO production in RAW264.7 cells[72][107].

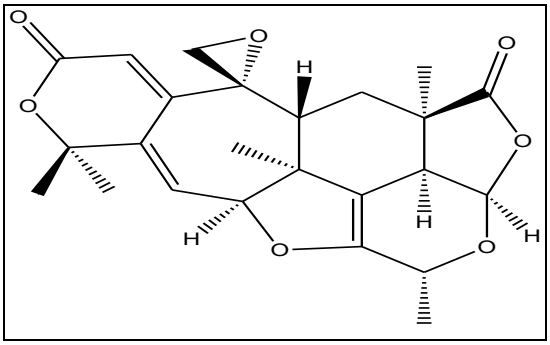
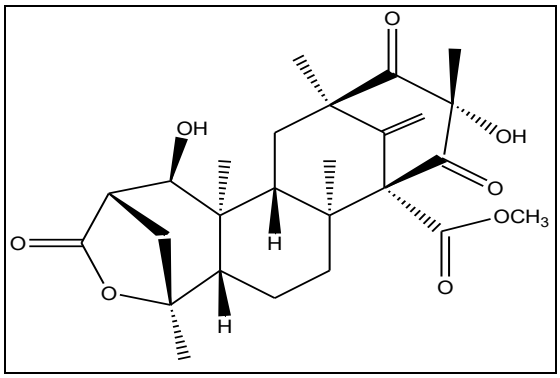
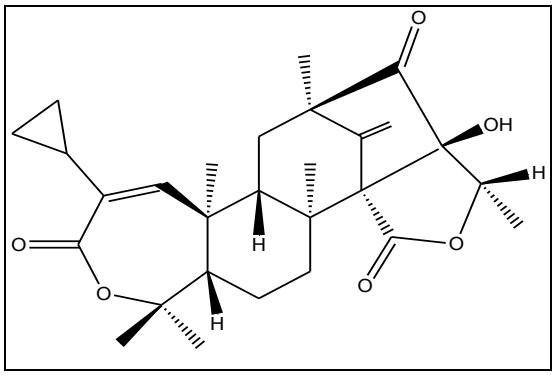


Fig 19: Amestolkolides A

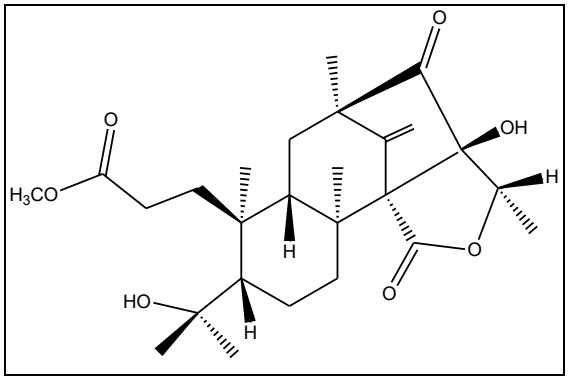
Brasilianoids A (Fig. 20a) from the marine-derived fungus *Penicillium brasilianum* WZXY-m122-9, isolated from an unidentified sponge, significantly dose-dependently induced the expression of filaggrin and caspase-14 in HaCaT (human keratinocyte) cells. As filaggrin plays a crucial role as a natural moisturising factor in maintaining the skin's moisture barrier, this suggests its potential as a cosmeceutical ingredient for skin moisturization in the cosmetic industry[108]. Brasilianoid C (Fig. 20 b), brasilianoid D (Fig. 20 c), and brasilianoid F (Fig. 20 d)inhibit the DNA expression of the HBV virus in HepG2.2.15 0 cell line[109].

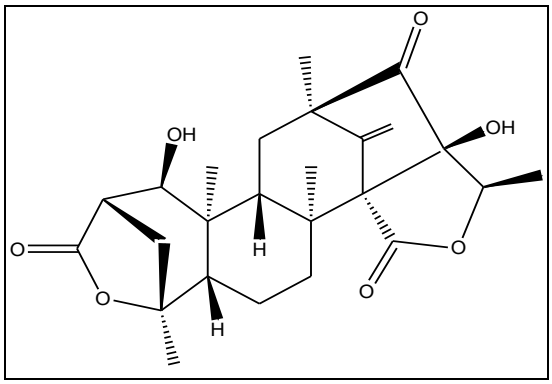


(a)



(b)

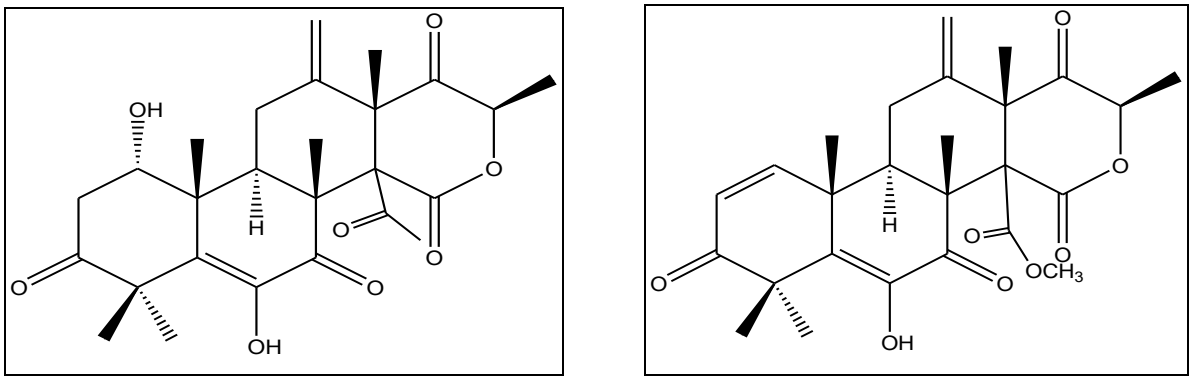




(c) (d)

Fig 20: (a) Brasilianoids A (b) Brasilianoids C (c) Brasilianoids D (d) Brasilianoids F

Terretonins E & F (Fig. 21 a-b) were isolated from the culture extract of the marine-derived fungus *Aspergillus insuetus*, obtained from the marine sponge *Petrosia ficiformis* collected in the Mediterranean Sea. Among terreustoxins A-M, terreustoxin C and terretonin have been reported for their proliferation inhibitory activity on Con A-induced murine T cells[110]. Furanasperterterpenes A and 11-acetoxy-terretonin E show cytotoxic activity in the 3T3-L1 adipocyte cell test system, where Furanasperterterpenes A was less effective than 11-acetoxy-terretonin E[11].



(a) (b)

Fig 21: (a) Terretonins E (b) Terretonins F

Andrastin A, andrastin B, citreohybridonol, 3-deacetylcitreohybridonol, 6α-hydroxyandrastin B, dihydrocitreohybridonol, (Fig. 22a- f) conidiogenone C, conidiogenone I was tested for their inhibitory effect against NO production in RAW 264.7 macrophages. Other than conidiogenone C and conidiogenone I compound, exhibited inhibition ranging from 60% to 90% while conidiogenone C showed notable inhibition rates of 63% and 91%. All compounds other than conidiogenone C showed a notable cytotoxic effect, while conidiogenone C had a weak cytotoxic effect, which is confirmed by MTT assay[111]. Andrastone A, a distinct andrastin bearing an unusual cyclopentane-1,3-dione, along with 16-epi-citreohybriddione A & citreohybriddione A isolated from a marine fungi *Penicillium allii-sativi,* showing cytotoxic activity on various cell lines such as HepG2 by the MTT assay method. Among the three compounds, andrastone A showed a selective effect on HepG2 cells via Caspase and RXRα pathways[112]. 12-epi-aspertetranone D shows 41% inhibition of colony formation of 22Rv1 prostate cancer cells in MTT assay(12). Andrastone C and andrastone B show antimicrobial action against methicillin-resistant *S. aureus* (MRSA*), E. coli,* & *C. albicans*[113].

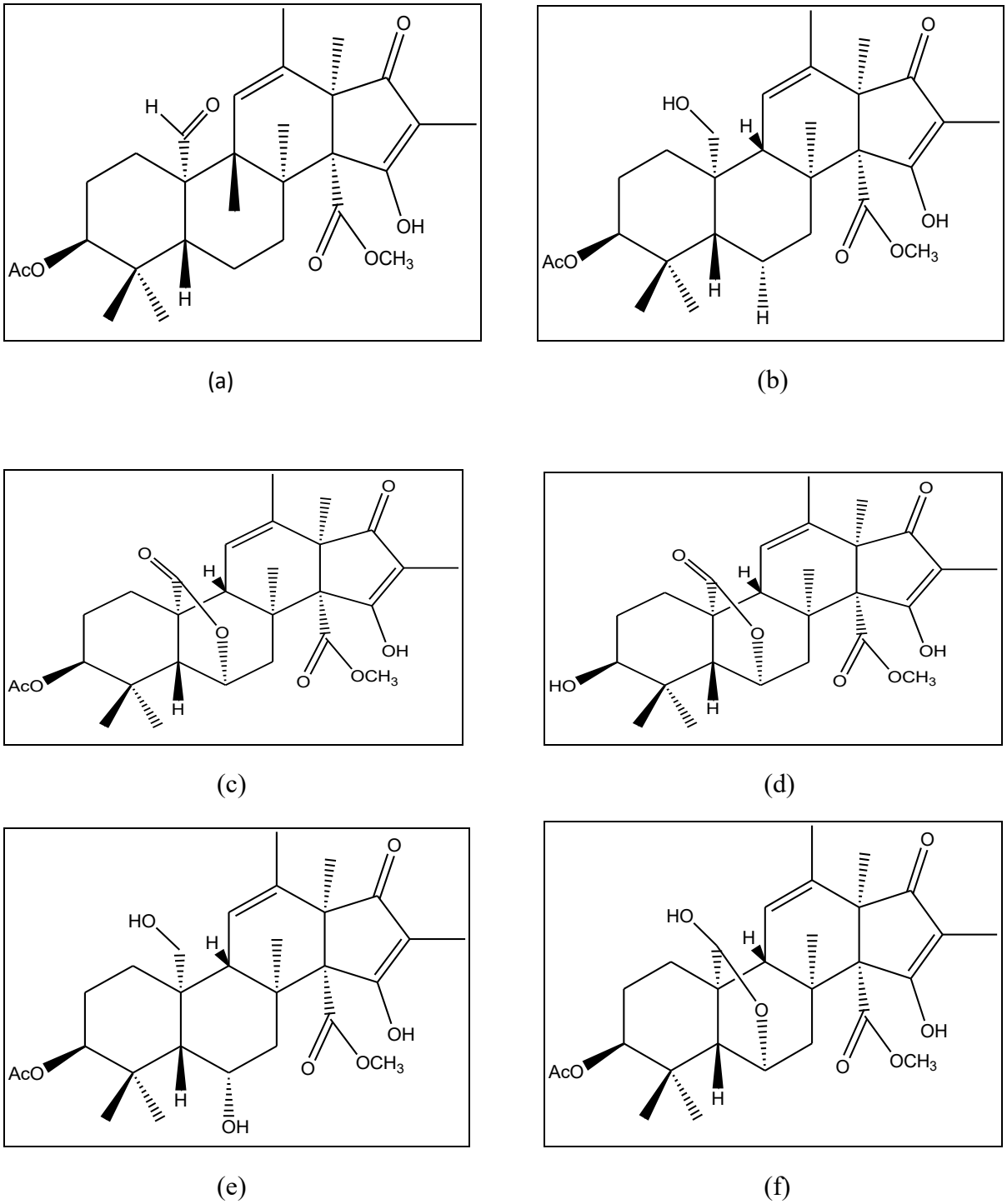


Fig 22: (a) Andrastin A (b) Andrastin B (c) Citreohybridonol (d) 3-deacetylcitreohybridonol (e) 6α-hydroxyandrastin B (f) Dihydrocitreohybridonol

Simpterpenoid A (Fig 23), a drimane merosesquiterpene linked to rearranged 3,5-dimethylorsellinic acid, containing a highly functionalized cyclohexadiene moiety with gem-propane-1,2-dione and methylformate groups was isolated from the culture extract of *Penicillium simplicissimum* MA-332, obtained from the rhizospheric soil of the marine mangrove plant *Bruguiera sexangula var. rhynchopetala*, and exhibited potent in vitro inhibitory activity against influenza neuraminidase[8].

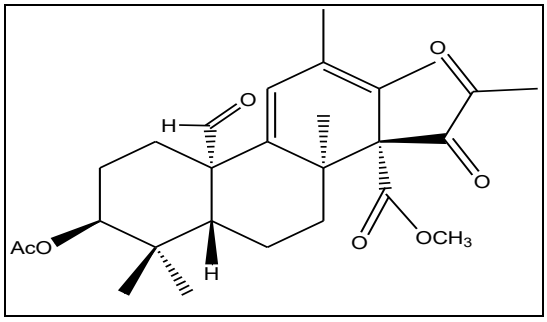


Fig 23: Simpterpenoid A

Aspertetranones A–D (Fig 24 a-d), highly oxygenated rearranged drimane merosesquiterpenes linked to an isochromone moiety, and isolated from the culture extract of the endophytic fungus Aspergillus sp. ZL0-1b14, obtained from the marine green alga of the genus Enteromorpha, have been shown to display weak inhibitory effects on TNF-α and NO production in LPS-stimulated RAW264.7 macrophages[114].

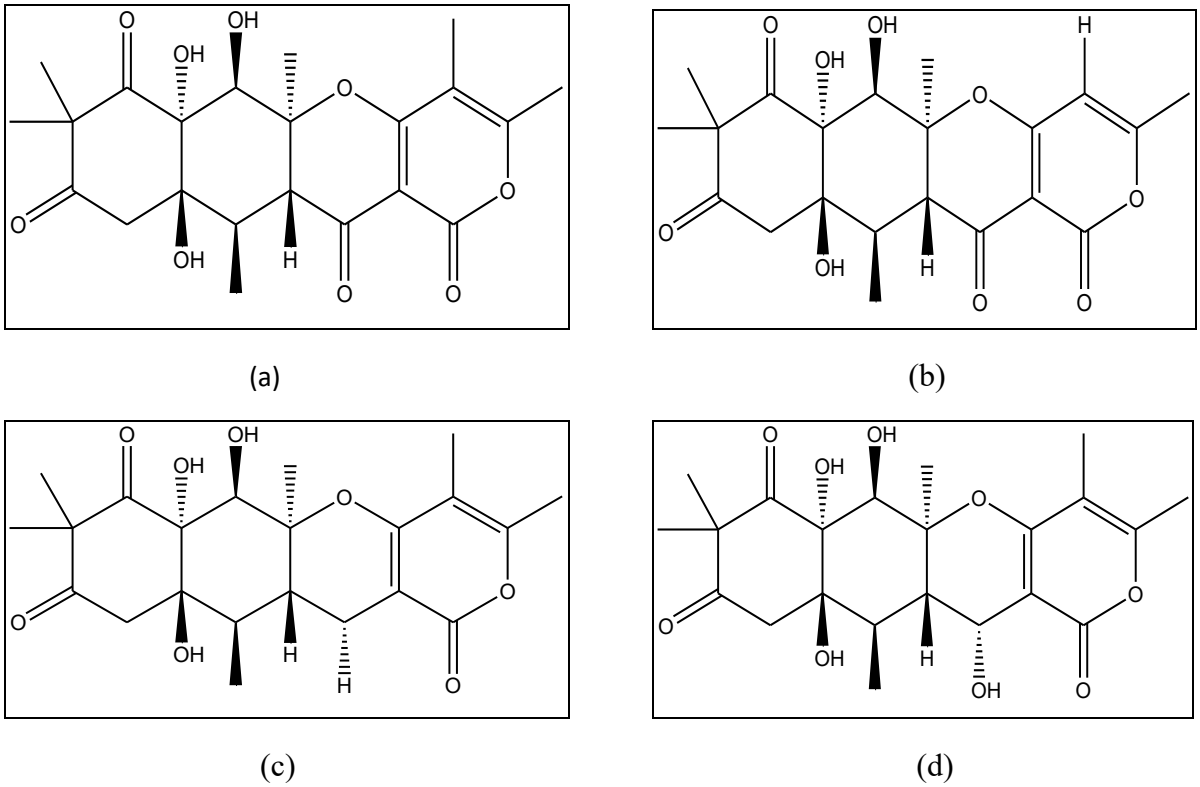


Fig 24: (a) Aspertetranones A (b) Aspertetranones B (c) Aspertetranones C (d) Aspertetranones D

Meroditerpenoids, a class of naturally occurring compounds, can be divided into three main categories: (i) diterpenes linked with 3,5-dimethylorsellinic acid, (ii) diterpenes linked with polyketides, and (iii) diterpenes linked with indole derivatives[115][116].

Diterpenes Linked to 3,5-Dimethylorsellinic Acid, such as terreusterpenes A–B (Fig. 25a- b), isolated from the culture extract of *Aspergillus terreus*, obtained from the inner part of the soft coral *Sarcophyton subviride,* which is collected from the Xisha Island, China, exhibited potent inhibitory activity against β-site amyloid precursor protein-cleaving enzyme 1 (BACE 1) [116].

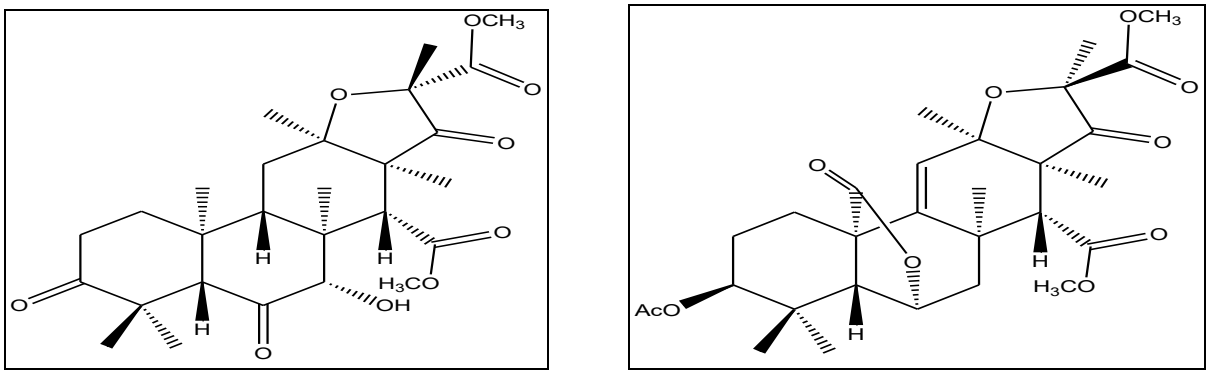


Fig 25: (a) Terreusterpenes A (b) Terreusterpenes B

Indole-diterpenoids, penerpene A and B, obtained from a marine fungus Penicillium species KFD28, which shows potent inhibitory action towards protein tyrosine phosphatases[117].

Prenylated hydroquinones & chrysogenester found in *Penicillium chrysogenum* on docking analysis showed that chrysogenester binds to PPAR-γ in the same manner as the natural PPAR-γ agonist amorfrutin B. It inhibits the phosphorylation of the subunit of the NF-κB p65 and suppresses the expression of the iNOS, NO, COX-2, TNF-α, IL-1β, & IL-6 pro-inflammatory mediators by activation of PPAR-γ, followed by downregulation of the NF-κB signalling pathway, which reduces the effect of pro-inflammatory mediators[5]. Antroquinonol, a ubiquinone derivative derived from the medicinal mushroom Antrodia camphorata, inhibits colon cancer cell growth by reducing stem cell-like characteristics through modulation of the PI3K/AKT/β-catenin signalling pathway[118]. Varitriol and varixanthone have been isolated from the marine fungi *Emericella variecolor,* the former being shown to possess 100-fold increased potency over the mean toxicity towards cancer cell lines[119], while the latter is a strong antimicrobial against *E. coli, B. subtillis, Proteus sp., S. aureus,* and has less potential toward *E. faecalis*[13]. Rel (6aS,10aR)- Δ9-tetrahydrocannabiorcolic acid B are isolated from the endophytic fungus Pseudocosmospora species Bm 1-1 from *Acanthus ebracteatus* Vahl, showed significant cytotoxic activity on the HL60 cell line[120].

5. Conclusion

This review includes an overview of meroterpenes with their source, structures, and detailed pharmacological potential, along with biological synthesis that we hope will underpin future work on this class of secondary metabolites. This review mainly focuses on the major phytoconstituents functioning as meroterpenes along with their pharmacological effects. The article includes major phytoconstituents such as ubiquinone-10, α-tocopherol, and austinol, which are known for their potent therapeutic actions. Meroterpenoids exhibit anti-neoplastic, anti-microbial, antifungal, antimalarial, & anti-inflammatory actions. A few illustrious meroterpenoids include antidepressant hyperforin, anticancer vinblastine and vincristine, psychoactive drug tetrahydrocannabinol, and immunosuppressant mycophenolic acid. The phytoconstituents of this category, meroterpene, have huge potential. The detailed information presented in this review on the pharmacognostical features, phytochemical studies, various pharmacological activities, and the uses of phytoconstituents are included in the class of

meroterpene. Therefore, with appropriate knowledge, meroterpenes could be promoted to be used for the welfare of local people and industry, especially the pharmaceutical industry, and it may heighten food security, health security, environmental security, and economic security.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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