Efficacy of some natural compounds as antifungal agents

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ABSTRACT

Natural sources have been important for the development of new active molecules for many years. Various small molecules with unique chemical skeleton and potent bioactivities were discovered through various sources like plants, marine products, and microorganisms, etc., which are considered as very important part of the nature. A number of potent antifungals have been originated from various natural sources. This account describes structure and activities of selected agents isolated from various natural sources.

Key words: Antifungals, biological activities, chemical structure, natural sources

INTRODUCTION

In the immunological comprised patients single mold spore can initiate the deadly process. Patients on chemotherapy or immunosuppression, imposed for organ transplantation and those with chronic diseases such as AIDs, recurrent infection, cystic fibrosis, and diabetes are also at risk.[1,2] One of the factors aiding the spread of fungal disease has been the widespread use of broad-spectrum antibiotics, which eliminate or decrease the nonpathogenic bacterial populations that normally compete with fungi. Another has been the increased number of individuals with reduced immune responses caused by the acquired immunodeficiency syndrome or by the action of immunosuppressant drugs, or cancer chemotherapeutic agents. Besides their increasing frequency, fungal infections are still associated with an unacceptably high mortality, up to 40% in bloodstream infections caused by *Candida albicans*[3] and more than 50% in invasive aspergillosis.[4] The most commonly recognized causes of opportunistic invasive fungal infections (IFIs) traditionally are *C. albicans*, *Cryptococcus neoformans*, and *Aspergillus fumigates*.[5] Along with the widespread use of antifungal prophylaxis, the epidemiology of IFIs has shifted toward non-*albicans Candida*, non-*fumigatus Aspergillus*, opportunistic yeast-like fungi (e.g. *Trichosporon* and *Rhodotorula* spp.), zygomycetes, and hyaline moulds (e.g. *Fusarium* and *Scedosporium* spp.).[6] The limitations of current antifungal drugs, increased incidence of systemic fungal infections, and rapid development of drug resistance have highlighted the need for the discovery of new antifungal agents, preferably with novel mechanism of action.[7,8] While synthetic efforts remain the mainstream in antifungal drug discovery, as evidenced by the fact that 18 out of 23 antifungal drugs approved from 1980 to 2002 are synthetic (83% belongs to single azole class), unique antifungal natural products have demonstrated remarkable success in this regard.[9] For example, two major drug classes currently in use (amphotericin B, the gold standard and armamentarium, and the lipopeptide caspofungin) are the important antifungal drugs approved in recent years, are derived from natural products.[10] Natural products provide opportunities for new drug leads. This account describes structure and activities of selected agents isolated from various natural sources with the aim to examine the recent developments toward discovering novel antifungal drugs of natural origin. However, we have excluded certain compounds that are often termed "nuisance" compounds, because they show a variety of biological activities.

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ANTIFUNGALS FROM NATURAL SOURCES

Phytochemical analysis of *P. costaricense* parts leads to the isolation and identification of δ-toco trienol, β-sitosterol, and other active constituents. Cinnamodial (1) and cinnamosmolide (**2**) were found to have high activity against *Alternaria alternate* (MIC=3.9μg/ml), *C. albicans*, and *Wangeiella dermatides* (MIC=15.6μg/ml). Compound (**2**) showed less potent antifungal activities than (1) but was more effective against *C. albicans* CNIA (MIC = 23.4μg/ml).[11]

Twentyfive natural products mainly halogenated furanones were isolated from the temperate red algae *Delisea pulchra*. The activity was found to be very strong against fungus *P. oxalis* for 18 compounds, 10 compounds (3-12) maintained strong activity at 0.1 μg/ml while two compounds 10 and 12 have shown activities comparable to miconazole [Figure 1].[12]

Carneic acids (13, 14)isolated as major constituents of the stromata of hypoxylon carneum have shown moderate antifungal activity against various fungal infections.[13] Hopeanolin (15), an unusual resveratral trimer with an *ortho*-quinone nucleus, from stem bark of *Hopea exalata* demonstrated very good antifungal activity [Figure 2]. This plant has six known stibenoids, shoremphinol (16), vaticanol (17), α-viniferin (18), pauciflorol A (19), vaticanol A (20), and *trans*-3,5,4′-trihydroxystilbene 2-C-glucoside (21) have shown antifungal activities in the MIC value range 0.1–22.5 μg/ml.[14] Two new macrolides, swinholides I (22) and hurghadolide A (23, 24) of which hurghadolide A possesses an unprecendent asymmetric 42-membered dilactone moiety were reported. Compounds 23 and 24 have been demonstrated to be active against *C. albicans*.[15]

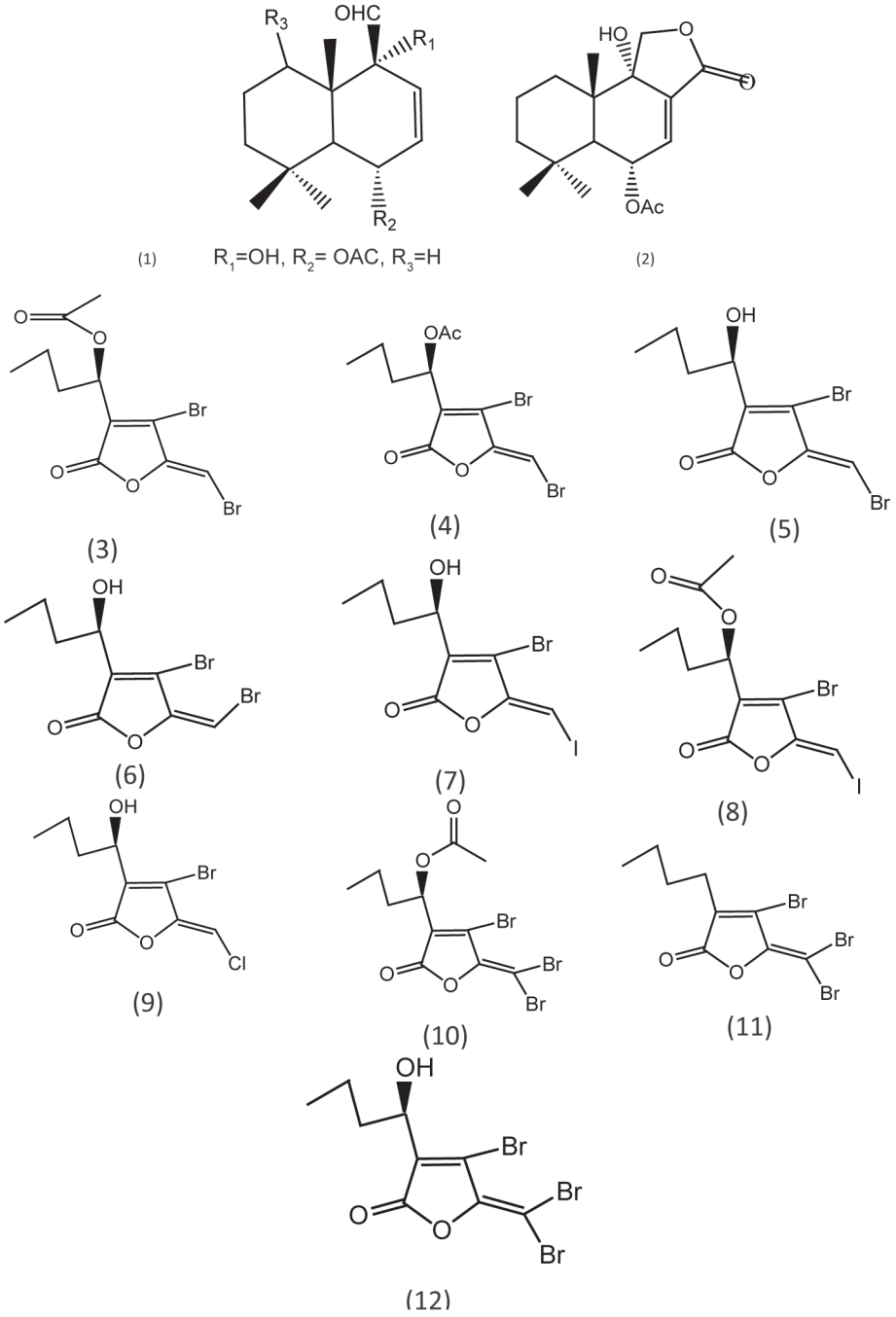

Figure 1: Halogenated furanones obtained from algae as antifungal agents

Marine-derived macrolides latrunculins A and B of the red sea sponge *Negombata magnifica* are the marine products. Latrunculin B (25) together with latrunculin T (26) was isolated and certain semisynthetic analogs (27, 28, 29, and 30) were reported to have potent activity against *C. albicans*, the presence of αβ lactones in the ring is responsible for the activities.[16]

Five new *ent*-labdane diterpenoids, 3-*o*- β D-glucopyranosyl-14,19-dideoxyandrographolide (31), 14-deoxy-17-hydroxy-andrographolide (32), 19-*o*-[β-D-glucopyranosylandrographolide (33), 12-s-hydroxy andrographolide (34), and andrographatoside (35) were isolated from the aerial part of *andrographis paniculata* and all compounds were active against various fungal strains [Figure 3].[17]

Three new *N*-methyl-4-hydroxy-2-pyridinone analogs, 6-epi-oxysporidinone (36), were isolated from the fungus *Fusarium oxysporum* (N17B). The compound showed selective fungistatic activity against *Aspergillus fumigatus*.[18] Four dialkylresorcinols (37-40) were isolated from the liquid culture of *Pseudomonas* sp. Ki19. These compounds were reported to inhibit *A. fumigatus* and *Fusarium culmorum* at 50 μg/ml.[19] Three linear sesquiterpene lactones, anthecotulide (41), hydroxyl anthecotulide (42), and acetoxyanthecotulide (43) were isolated from aerial parts of *Anthemis auriculata*.[20]

Two new azapilones named rotiorinols (44, 45) and stereoisomer (-)-rotiorin (46), and a known compound rubrotiorin (47) were isolated from the fungus *Chaetorium cupreum* CC3003. Compounds exhibited activity against *C. albicans* with IC_{50} values of 10.5, 16.7, 24.3, and 0.6, respectively [Figure 4].[21] Crysotriones A (48) and B (49), two new 2-acylcyclopentene-1,3-dione derivatives, were isolated from the fruiting bodied of the Basidiomycete Hygrophorus chrysodon found in mushrooms have shown activity against *Fusarium verticillioides*.[22] Two new peptaibols, septocylindrin A and septocylindrin B, were obtained from the fungus *Septocylindrium* sp.[23] A new marine -derived macrolides as neopeltolide (50) were reported to have activity against fungal pathogen *C. albicans* with MIC of 0.62 μg/ml.[24] The natural product dihydroferulic acid (DFA, 51) and the six synthesized derivatives of it were examined for antifungal activities. Test fungi included *Saccharomyces cerevisiae*, *A. fumigatus*, and *A. flavus*.[25] Six new minor dammarane triterpenoids (52, 53) were isolated from the roots of *Gentiana rigescens*. These are the glycosides and have shown antifungal activity against the pathogen *Glomerella cingulata*.[26]

Four solanapyrone analogs (solanapyrones, 54–57) were reported from an unidentified fungicolous fungus and showed activity against *A. flavus* and *F. verticillioides*, and *C. albicans*.[27] Two volatile benzaldehyde derivatives (58, 59) were isolated from a

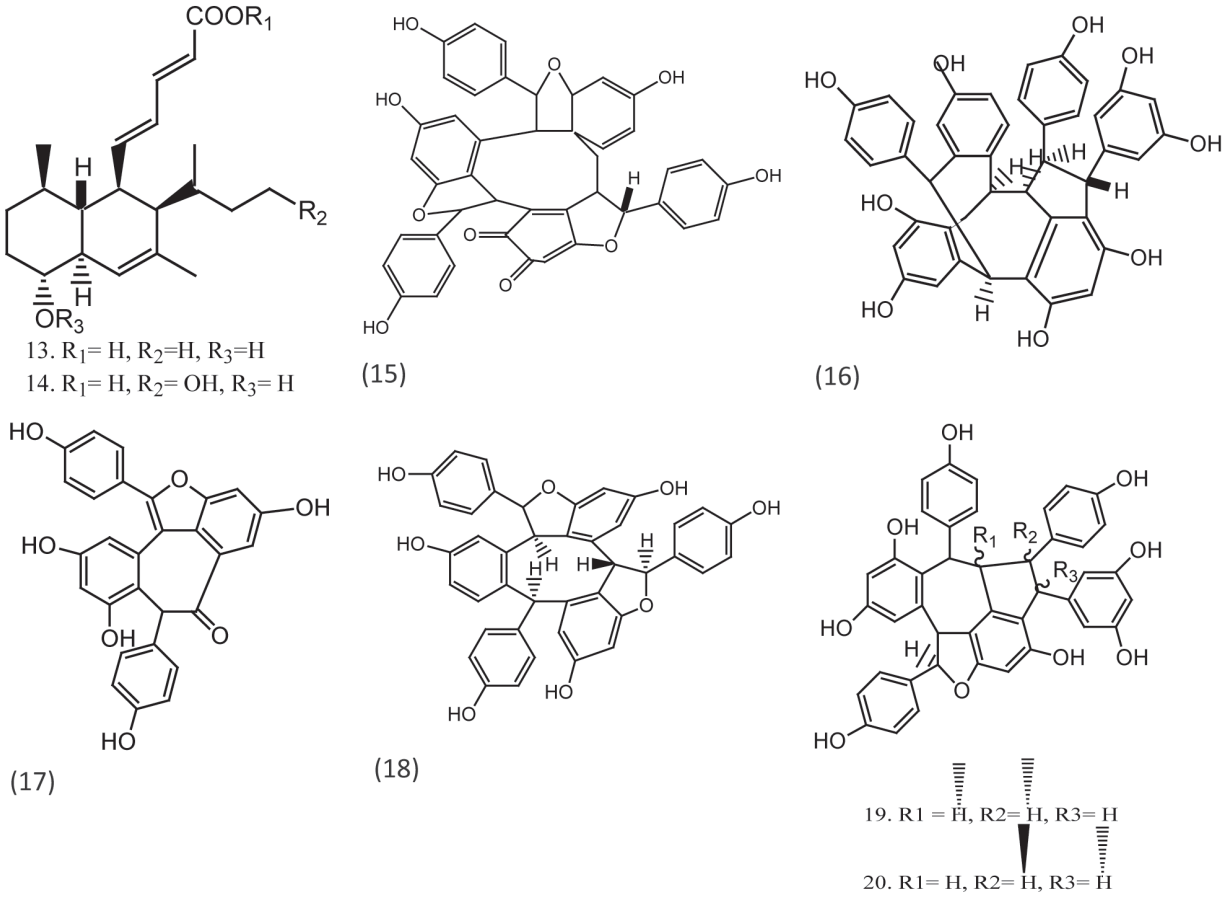

Figure 2: Compounds with quinone, furan nucleus

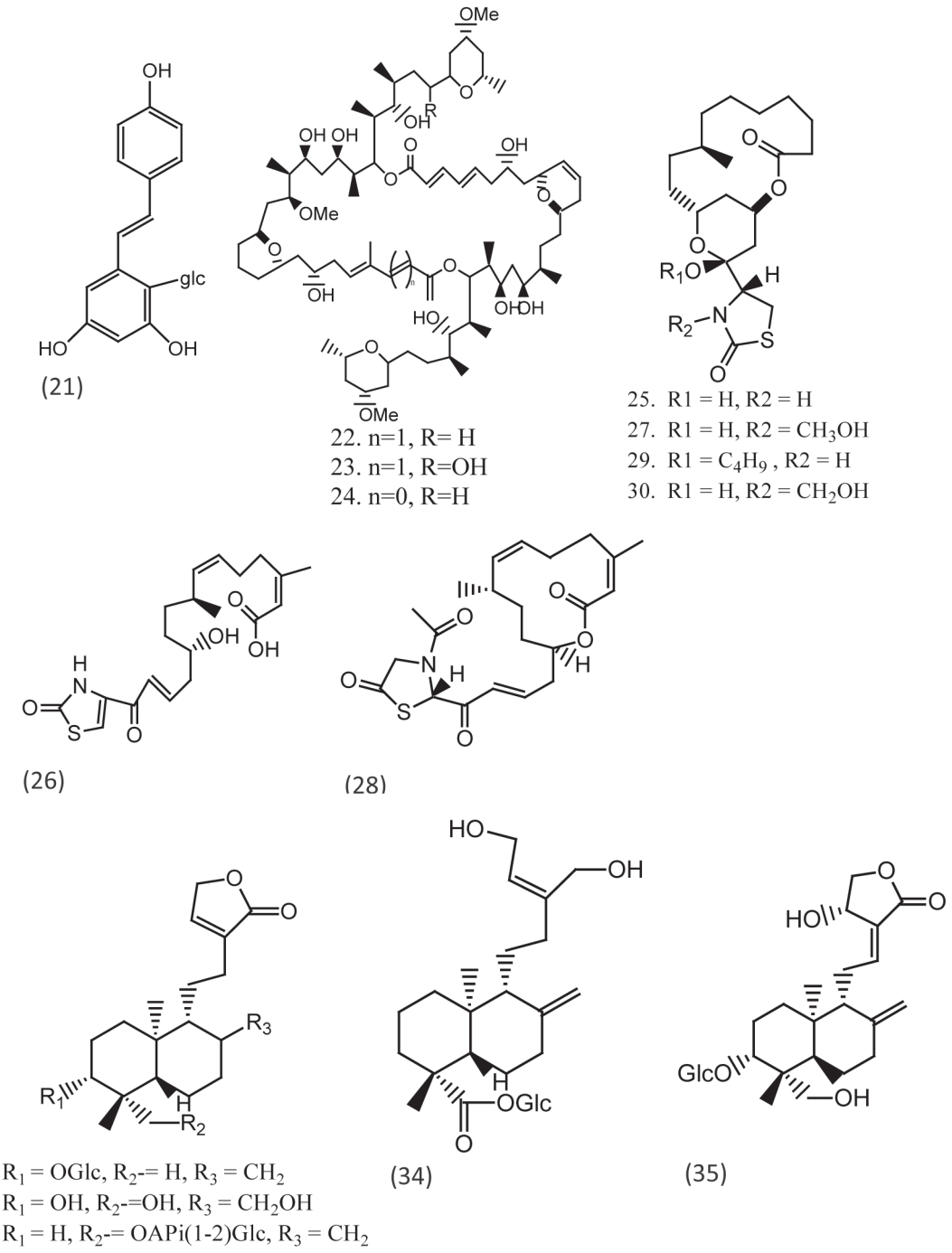

Figure 3: Compounds with thiazoles and macrolide structures

Basidiomycete Sacodontia crosea found to be active against various pathogenic fungi. The aerial part of *Centaurea pullata* resulted into compounds (60, 61) which is reported to have potent activity [Figure 5].[28]

Leaves of *Piper scutifolium* yielded two new isobutyl amides, scutifolium A and scutifoliamide B, which were reported as fungistatics.[29] Compound phaeosphenone (62, 63) was isolated from *Staphylococcus aureus* and inhibited the growth of *C. albicans* with an MIC of 8 μg/ml.[30] 4-Terpenyl cannabinolate (64) which is isolated from *Cannabis sativa* have shown activity against *C. albicans* ATCC 90028 with an IC_{50} value of 8.5 μg/ml.[31] 1α,5α-Dioxy-11 α-hydroxyurs-12-en-3-one (65) and urasane triterpenoid (66) exhibited modest activity against *E. coli*, *C. albicans* with the MIC 31.9μM/L and 36.3μM/L, respectively.[32]

Pestafolide A (67), a new reduced spiro azaphilone derivative, and pestaphthalides A (68) and B (69) were isolated from *Pestalotiopsis foedan.* These compounds have shown activity against *C. albicans* (ATCC 10231), *Geotrichum candidum*, and *A. fumigates* (ATCC 10894).[33]

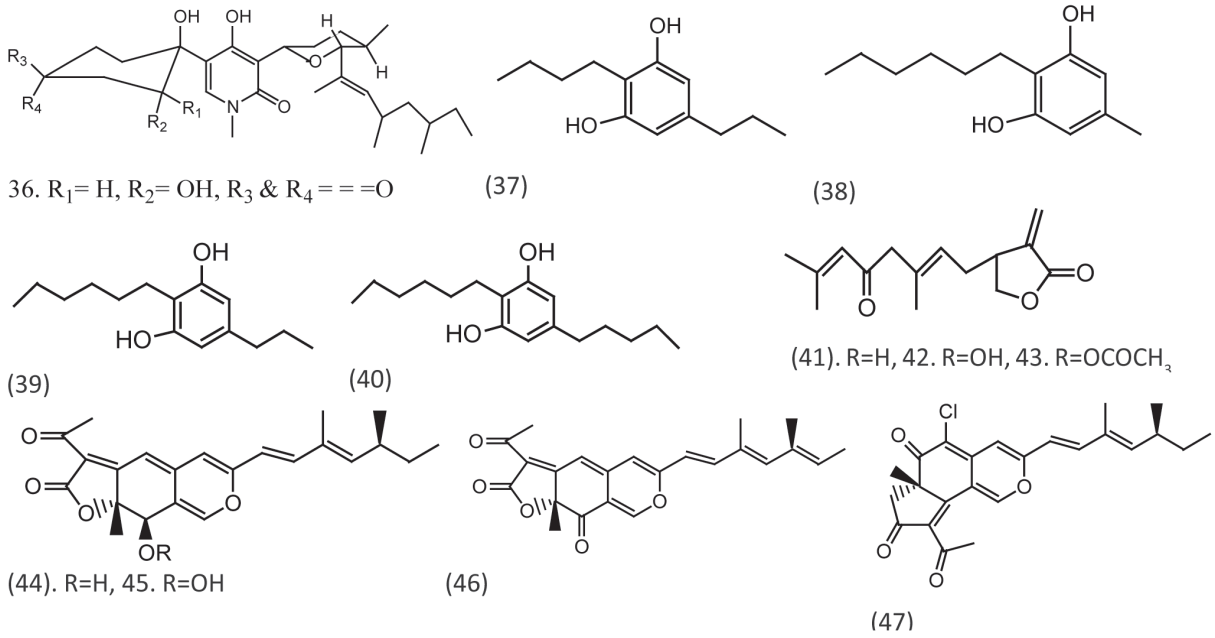

Figure 4: Compounds reported with antifungal activity from different chemical categories

Lipophilic extracts of the stem bark of *Buddleja globosa* were found to have antifungal activity at 125 ìg/ml against three dermatophytic fungal species *Trichophyton rubrum*, *Tricophyton interdigitale*, and *Epidermophyton floccosum*. The known compounds were the diterpene buddlejone (70), the bisditerpene maytenone, and the two sesquiterpenes buddledin A and buddledin B, while the novel compound was characterized as the diterpene deoxybuddlejone (71).[34]

From witches' broom diseased bamboo, *Phyllostachys bambusoides*, *N-p*-coumaroylserotonin, and *N*-feruloylserotonin were isolated. *N-p*-Coumaroylserotonin possesses antifungal activity against *Aciculosporium take*, the causal agent of witches' broom of bamboo. A new antifungal substance against rice blast fungus (*Pyricularia oryzae*), 5-(8'-*Z*-heptadecenyl) resorcinol, was isolated from etiolated rice seedlings together with a mixture of its homologs with C_{13}, C_{15}, and C_{17} saturated alkyl chains. Its structure was determined by ^{1}NMR, ^{13}C-NMR, and MS spectra. Its ED_{50} was 40μg/ml.[35]

Two antifungal terpenoids, 6-isopropyltropolone β-glucoside and 5-(3-hydroxy-3-methyl-*trans*-1-butenyl)-6-isopropyl-tropolone β-glucoside, named by us cupressotropolone A and B, respectively, were isolated from the bark of Italian cypress, in response to infection by the fungus *Diplodia pinea* f. sp. *cupressi*. These tropolone glucosides inhibited *in vitro* germination of spores of *Diplodia pinea* f. sp. *cupressi, Seiridium cardinale, Alternaria alternate*, and *Verticillium dahliae*.[36] The antifungal activity of *Monilinia fructicola* from some naturally occurring pterocarpans were reported with ED50 values around 2×10^{-5} μg/ml.[37]

Secondary metabolites were extracted from culture filtrates of three isolates of the mycoparasitic biocontrol agent *Verticillium biguttatum*. The two antifungal metabolites were 1-[2',3'-dihydroxy-5'-(hydroxymethyl)phenyl]-3-methyl-but-2-ene (72) and 1-[2'-hydroxy-3'-methoxy-5'(hydroxymethyl) phenyl]-3-methyl-but-2-ene (72, 73). These new metabolites were named bigutol and methylbigutol, respectively. Low concentrations of each metabolite inhibited mycelial extension of *Rhizoctonia solani* (minimum inhibitory concentration 138 μg/ml). Production of these antifungals by *V. biguttatum* suggests that antibiosis may play a role during biocontrol by this mycoparasite, particularly of plant diseases caused by *R. solani*.[38] Four new phenalenone-type phytoalexins, named musanolones (74–77), have been isolated from infected rhizomes of banana plants (*Musa acuminata*; AAA cultivar Grand Nain). All the compounds have reported to have a strong inhibitory activity on the growth of the germination tube of *F. oxysporum* f. sp. *cubense* race 4.[39]

Bioassay-guided fractionation resulted in the isolation of four antifungal agents from the roots of *Cudrania cochinchinensis*. Two of these were new compounds, cudraxanthone S [1,3,5,6-tetrahydroxy-2-(1,1-dimethyl-2-propenyl)xanthone] (78) and cudraflavanone B (2′,4′,5,7-tetrahydroxy-6-prenylflavanone) (79). These compounds exhibited antifungal activities against *C. neoformans*, *A. fumigatus*, and *A. nidulans* (MICs , 2–8 μg/ml) [Figure 6].[40]

Vitis vinifera antimicrobial peptide (Vv-AMP1), isolated from *Vitis vinifera*, shown to encode a plant defensin. Recombinant Vv-AMP1 showed nonmorphogenic antifungal activity against a broad spectrum of fungi, probably altering the membrane permeability of the fungal pathogens. Recombinant Vv-AMP1 was extremely heat-stable and showed strong antifungal activity against a broad spectrum of plant pathogenic fungi, with very high levels of activity against the wilting disease causing pathogens *F. oxysporum* and *Verticillium dahliae*. The Vv-AMP1 peptide did not induce morphological changes on the treated

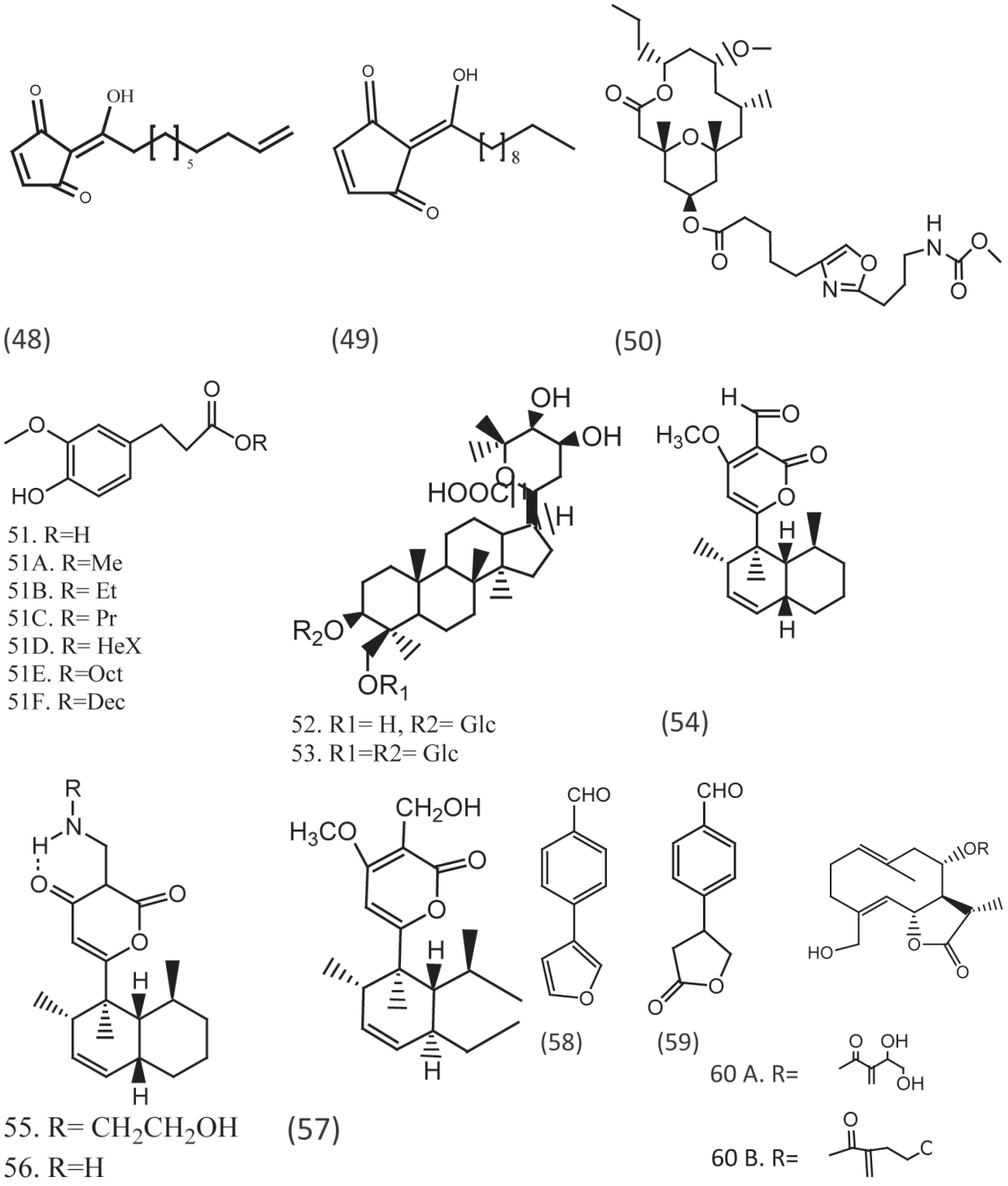

Figure 5: Compounds with remarkable antifungal activities

fungal hyphae, but instead strongly inhibited hyphal elongation. A propidium iodide uptake assay suggested that the inhibitory activity of Vv-AMP1 might be associated with altering the membrane permeability of the fungal membranes.[41, 42]

Dalbergia is a genus of trees, shrubs, and woody climbers widely distributed in tropical and sub- tropical regions. It possesses good antimicrobial activity.[43] Terminalia chebula plant was also reported to have antifungal activity.[44] Entada abysinnica (Fabaceae) leaves used by herbalists from the Lake Victoria region, Kenya. E. abysinnica could be a rich source of antimicrobial agents, especially antifungals (zones of inhibition were between 9.00 and 14.10 mm) against C. albicans, Sa. typhi, and St. aureus.[45] Butea monosperma has been reported for their antimicrobial activities.[46] Nyctanthes arbortristis have also shown great potential for antifungal activity.[47]

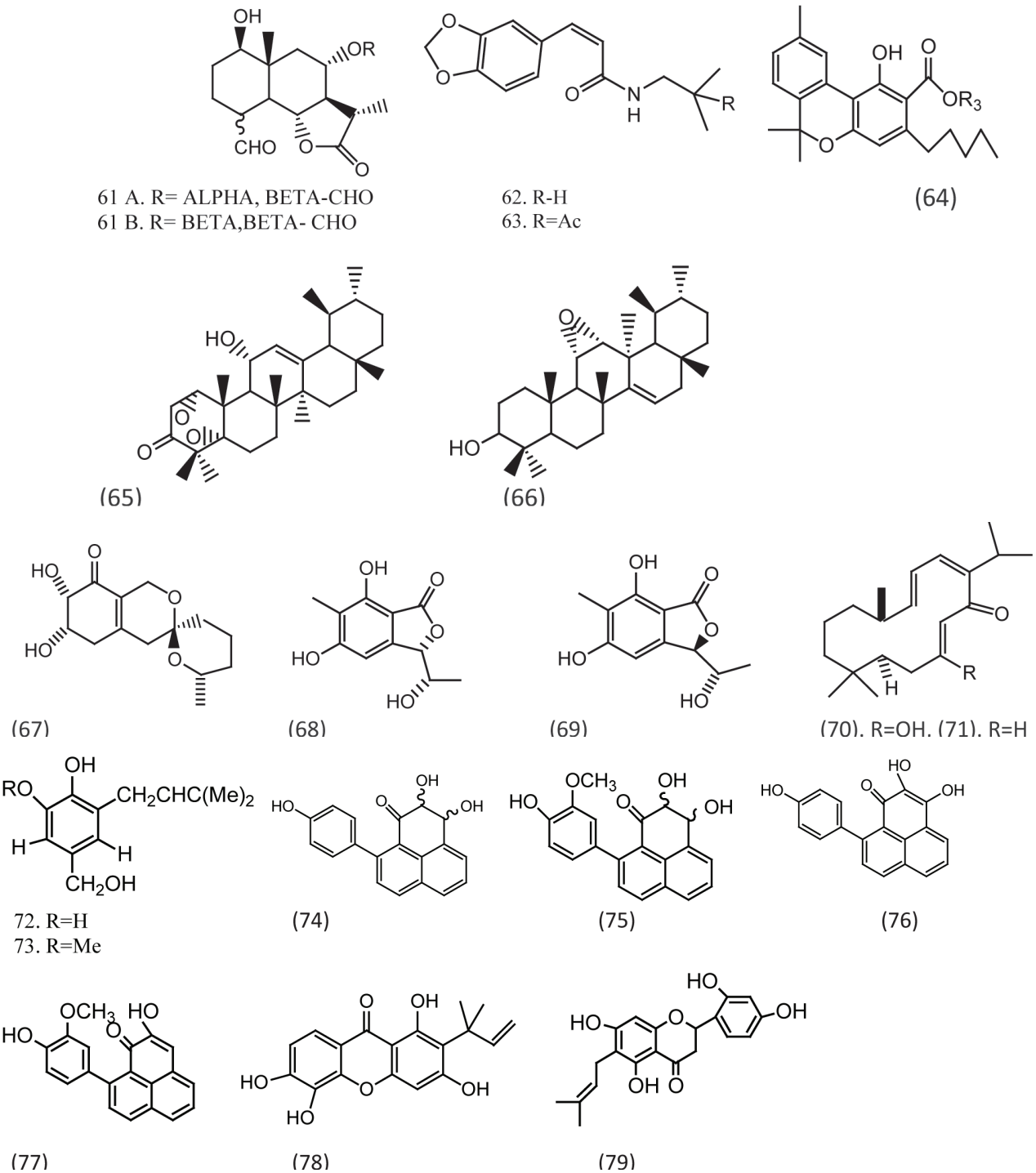

Figure 6: Compounds with good antifungal activities

CONCLUSION

A large number of metabolites from marine invertebrates, from plants, and from various other natural sources have been reported to inhibit the pathogenic fungi. These compounds represent a wide variety of structural classes ranging from cinnamodial sterols, furanones, quinines, and resorcinol to terpenoids in natural origin category. The spread of multidrug-resistant strains of fungus and the reduced number of drugs available make it necessary to discover new classes of antifungals and compounds that inhibit these resistant mechanisms. This has led to a search for therapeutic alternatives, particularly among medicinal plants and compounds isolated from them used for their empirically antifungal properties. In these natural sources, a series of molecules with antifungal activity against different strains of fungus have been found, which are of great importance

to humans and plants. It is hoped that other chemists will embark on the search for potent antifungal agents.

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