Secondary Metabolites from the Genus *Acremonium*

Subjects: Pharmacology & Pharmacy

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Acremonium fungi is one of the greatest and most complex genera in Hyphomycetes, comprising 130 species of marine and terrestrial sources. The past decades have witnessed substantial chemical and biological investigations on the diverse secondary metabolites from the *Acremonium* species. To date, over 600 compounds with abundant chemical types as well as a wide range of bioactivities have been obtained from this genus, attracting considerable attention from chemists and pharmacologists.

Acremonium secondary metabolites chemical structures bioactivities

1. Terpenoids

A total of 124 terpenoids have been reported in *Acremonium* fungi within the period 2016–2023, consisting of 31 sesquiterpenoids, 15 diterpenoids, 5 triterpenoids, and 78 meroterpenoids and miscellaneous types, while 99 compounds were found to have bioactivities. Remarkably, there are 20 new sesquiterpenoids, 11 new diterpenoids, 18 new meroterpenoids, and miscellaneous types.

1.1. Sesquiterpenoids

Several mophilane-type sesquiterpenoids, acremeremophilanes A–O (**1**–**15**), along with seven known analogues, were isolated from the deep-sea sediments derived from *Acremonium* sp. TVG-S004-0211 (**Figure 1**). Compounds **2**–**5** and **14** exhibited inhibition of lipopolysaccharide (LPS)-induced nitric oxide (NO) production in RAW 264.7 macrophages with IC_{50} values ranging from 8 to 45 μM [1].

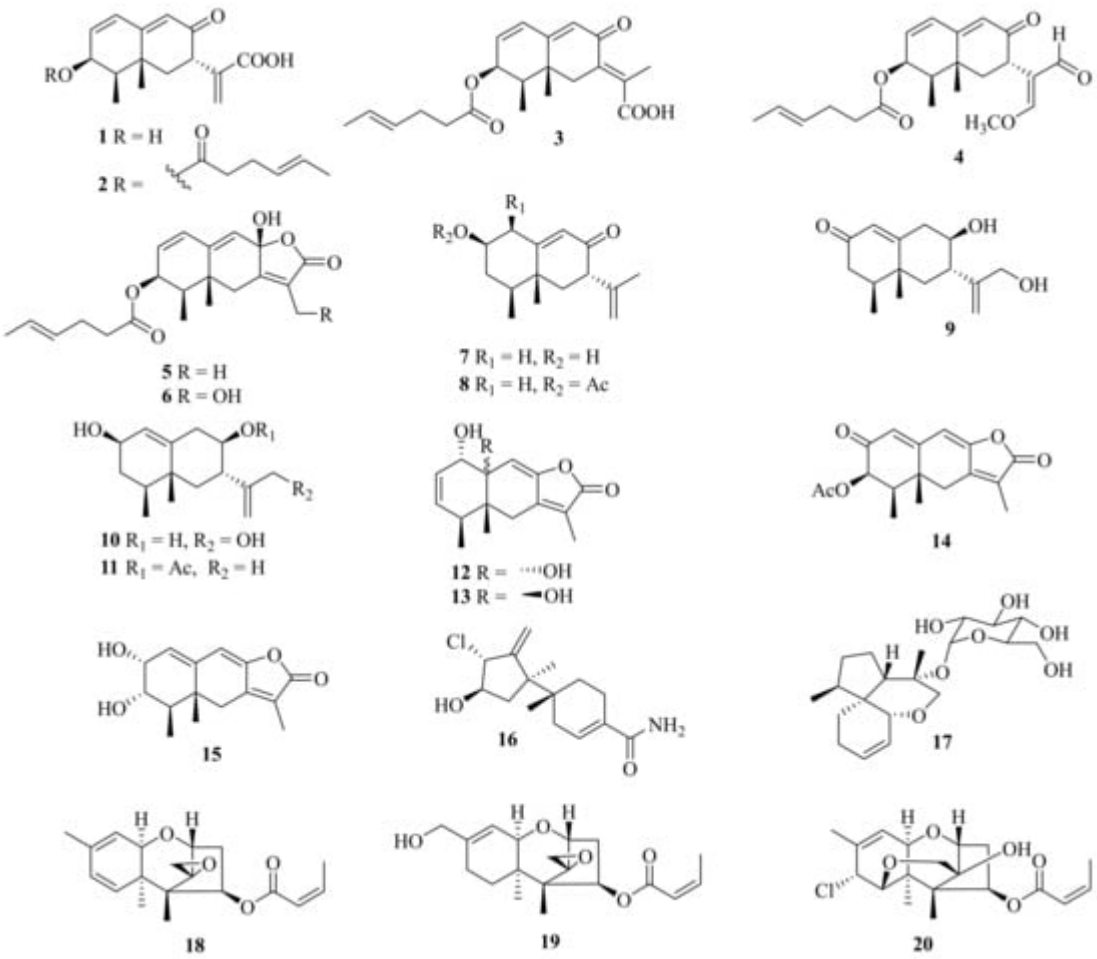

Figure 1. Chemical structures of sesquiterpenoids (**1**–**20**).

One new sesquiterpenoid, marinobazzanan (**16**), was isolated from marine sediment-derived *Acremonium* sp. CNQ-049, which showed an inhibition of cancer cell migration and invasion at non-toxic concentrations of 1, 2.5, and 5 μM by down-regulating transcription factors of Snail, Slug, and Twist. In addition, marinobazzananan reduced cell motility by down-regulating the expression level of KITENIN and by up-regulating the expression level of KAI 1, and it further reduced the number of metastatic nodules in the intraperitoneal xenograft mouse model [2]. Moreover, one new acorane-type sesquiterpene glycoside, isocordycepoloside A (**17**), was isolated from the fungus *Acremonium* sp. SF-7394 [3].

Meanwhile, three new trichothecenes, including two trichothecenes, 7-dehydro-8-dehydroxytrichothecinol B (**18**) and 8-deoxy-16-hydroxytrichothecinol B (**19**), along with one trichothecene analogue, 4-((*Z*)-but-2-enoyloxy)-8-chloro-12-hydroxy-7,13-epoxytrichothec-9-ene (**20**), and four known analogues, were isolated from the fungus *A. crotocinigenum* BCC 20012. Among them, the known compound trichothecin exerted the strongest antimalarial activity against *Plasmodium falciparum* K1 with an IC_{50} value of 0.05 mg/mL, and possessed cytotoxic activity against Vero cells with an IC_{50} value of 0.13 mg/mL [4].

1.2. Diterpenes

A chemical investigation of the marine-derived fungus *A. striatisporum* KMM 4401 resulted in the isolation of ten new diterpene glycosides, virescenosides Z9–Z18 (**21**–**30**), together with four known analogues [5] (**Figure 2**). One new diterpene, acrepseudoterin (**31**), was isolated from the fungus *Acremonium* sp. SF-7394. Acrepseudoterin inhibited the enzyme activity in a dose-dependent manner with an IC_{50} value of 22.8 ± 1.1 μM, which was identified as a competitive inhibitor of PTP1B [3].

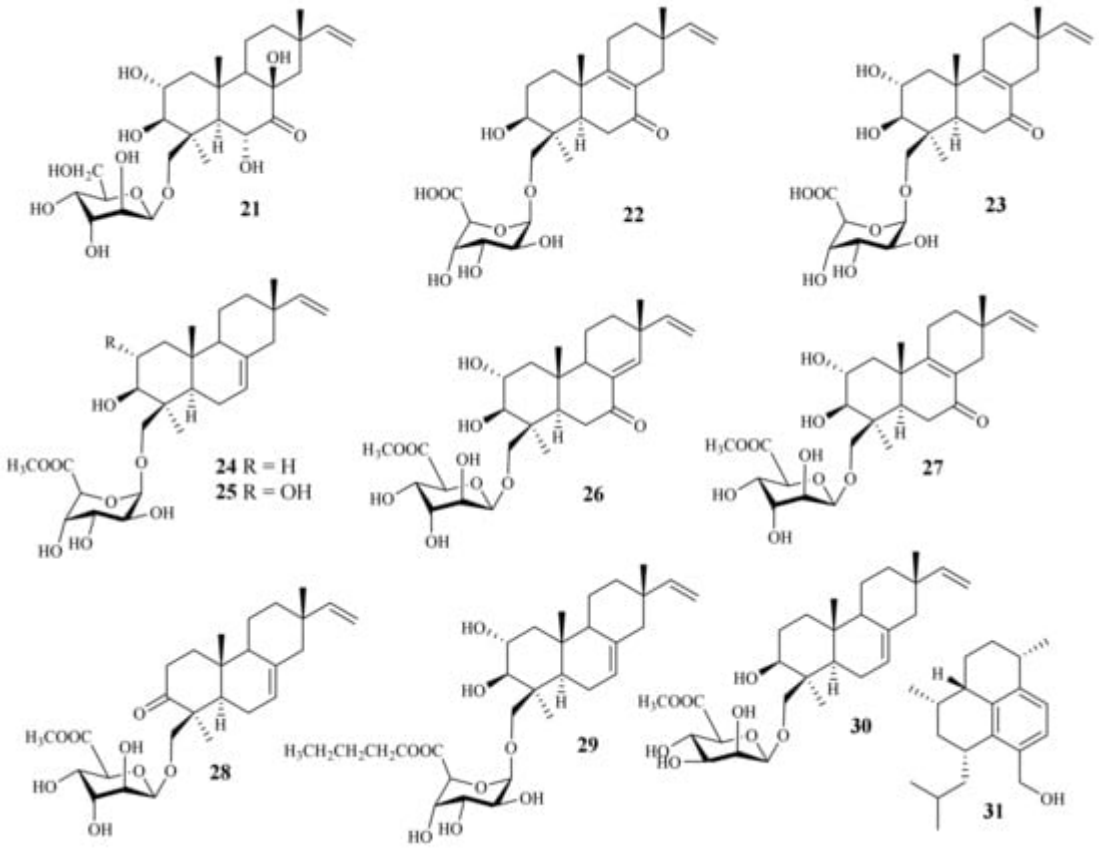

Figure 2. Chemical structures of sesquiterpenoids (**21**–**31**).

1.3. Meroterpenoids

Twenty-five ascochlorin derivatives, biosynthesized through the farnesylation of orsellinic acid [6], were obtained from the coral-derived *A. sclerotigenum* GXIMD 02501, including 13 new compounds, acremochlorins A–M (**32**–**44**) (**Figure 3**). Compounds **32** and **44**, two novel potent human dihydroorotate dehydrogenase (hDHODH) inhibitors, induced the apoptosis of triple-negative breast cancer (TNBC) cells by up-regulating the levels of cleaved-PARP1 and cleaved-caspase7, and further effectively inhibited tumor growth in a patient-derived TNBC xenograft model without significant weight loss or obvious toxicity in mice, showing higher safety than that of brequinar [7].

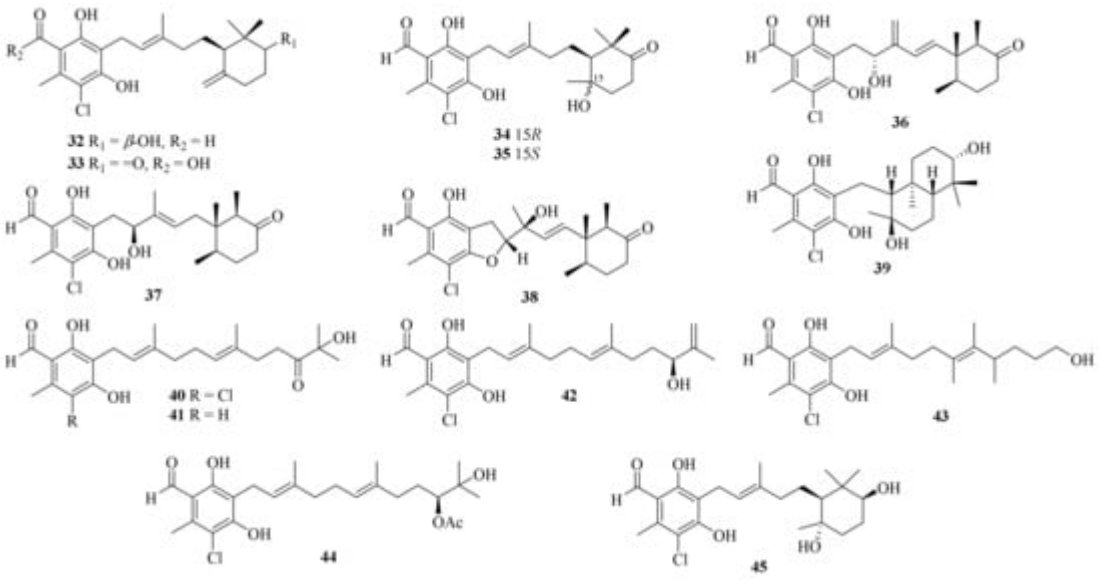

Figure 3. Chemical structures of meroterpenoids (**32**–**45**).

Meanwhile, ascofuranone and ascochlorin, two representative ascochlorin derivatives, were also reported as potential lead candidates for drug development targeting the hDHODH of cancer cells living under a tumor microenvironment [8]. Moreover, two known potential anti-tumor ascochlorins, 3-bromoascochlorin (BAS) and ilicicolin A (Ili-A), were also obtained from the coral-derived fungus *A. sclerotigenum* GXIMD 02501. BAS could induce the apoptosis, invasion, and migration of H446 and H69AR cells, and it further suppressed the tumor growth of a small cell lung cancer xenograft mouse model by inhibiting the mitogen-activated protein kinase (MAPK)/extracellular signal-regulated kinase (ERK) pathway [9]. Moreover, Ili-A showed efficacious activity against prostate cancer cells by abrogating EZH2/AR-mediated processes and demonstrated a synergistic anti-prostate cancer effect combined with enzalutamide in vivo, revealing a novel EZH2 inhibitor for the treatment of castration-resistant prostate cancer [10].

Ascofuranone and its derivatives, obtained from *A. egyptiacum*, were found as the first dual inhibitors of fumarate and oxygen respiration in *Echinococcus multilocularis* by targeting mitochondrial complexes II and III, suggesting potential lead compounds in the development of anthelminthic drugs [11]. One new ascochlorin, acremochlorin N (**45**), and a pair of new natural enantiomers, 3-phenylcyclopentane-1,2-diol (±-**46**) (**Figure 4**), together with nine known analogues were isolated from marine sediment-derived *A. furcatum* CS-280. All the isolates showed significant anti-*Vibrio* activities, especially against *Vibrio harveyi* and *V. alginolyticus*. Moreover, the presence of chlorine atoms in the ascochlorins could significantly enhance their antibacterial activity [12].

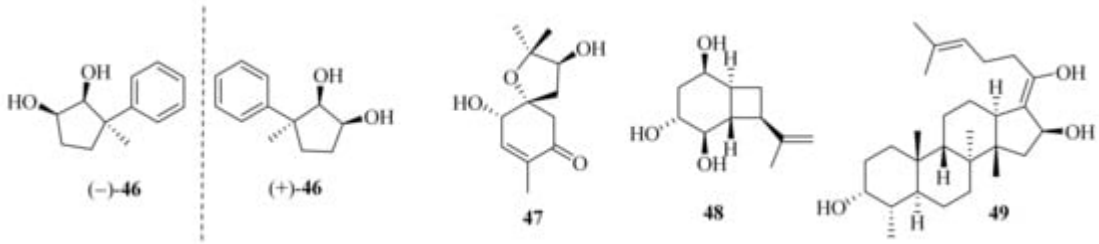

Figure 4. Chemical structures of miscellaneous terpenoids (**46**–**49**).

Meanwhile, four known ascochlorins, including ascochlorin, 10′-deoxy10′α-hydroxyascochlorin, 4′,5′-dihydro-4′-hydroxyascochlorin, and ascofuranone, were obtained from the sponge-derived *Acremonium* sp. IMB18-086. Ascochlorin and ascofuranone showed significant antibacterial activity against *Staphylococcus aureus*, methicillin-resistant *Staphylococcus aureus* (MRSA), *Bacillus subtilis*, and *Candida albicans*. Moreover, they showed significant cytotoxicity against A549 and/or HepG2 cell lines with IC_{50} values of 0.9–5.8 μM [13].

Acremine S (**47**) was isolated from the sponge *Mycale* sp. derived fungus *A. persicinum* KUFA 1007 and showed inhibitory activity against butyrylcholine esterase, which was three folds higher than that of galantamine [14]. Hexahydroacremonintriol (**48**), along with an analogue, acremonin A glucoside, were obtained from a tropical sinkhole derived from *A. masseei* CICY026. Both displayed insecticidal activity against *Myzus persicae* and/or *Rhopalosiphum padi* with settling inhibition ranging from 48% to 67% [15]. One new fusidic acid derivative, acremonidiol A (**49**), and three known analogs were obtained from the endophytic fungus *A. pilosum F47*. Among these, fusidic acid displayed a strong inhibitory effect on Gram-positive bacterium *S. aureus*, and the acetylation of the hydroxyl group at C-16 was crucial for antibacterial activity [16].

2. Peptides

A total of 45 peptides have been reported from *Acremonium* fungi during the period 2016–2023, including 33 new compounds, while 19 bioactive compounds were found.

2.1. Linear Peptides

One new linear peptide, acremopeptin (**50**), and a known one, adenopeptin, were obtained from the soil-derived fungus *Acremonium* sp. PF1450 [17]. Moreover, four new peptaibiotics, acremotins A–D (**51**–**54**) (**Figure 5**), along with a known peptaibiotic XR586 were isolated from the soil-derived fungus *A. persicinum* SC0105. Acremotins A–D showed strong inhibitory activity against Gram-positive bacteria, while the MIC values of acremotin D against *S. aureus* and MRSA were 12.5 and 6.25 μg/mL, respectively. Moreover, acremotins A–D and XR586 also showed cytotoxicity against three human cancer cell lines (A549, HeLa, and HepG2), with IC_{50} values ranging from 1.2 to 21.6 μM [18].

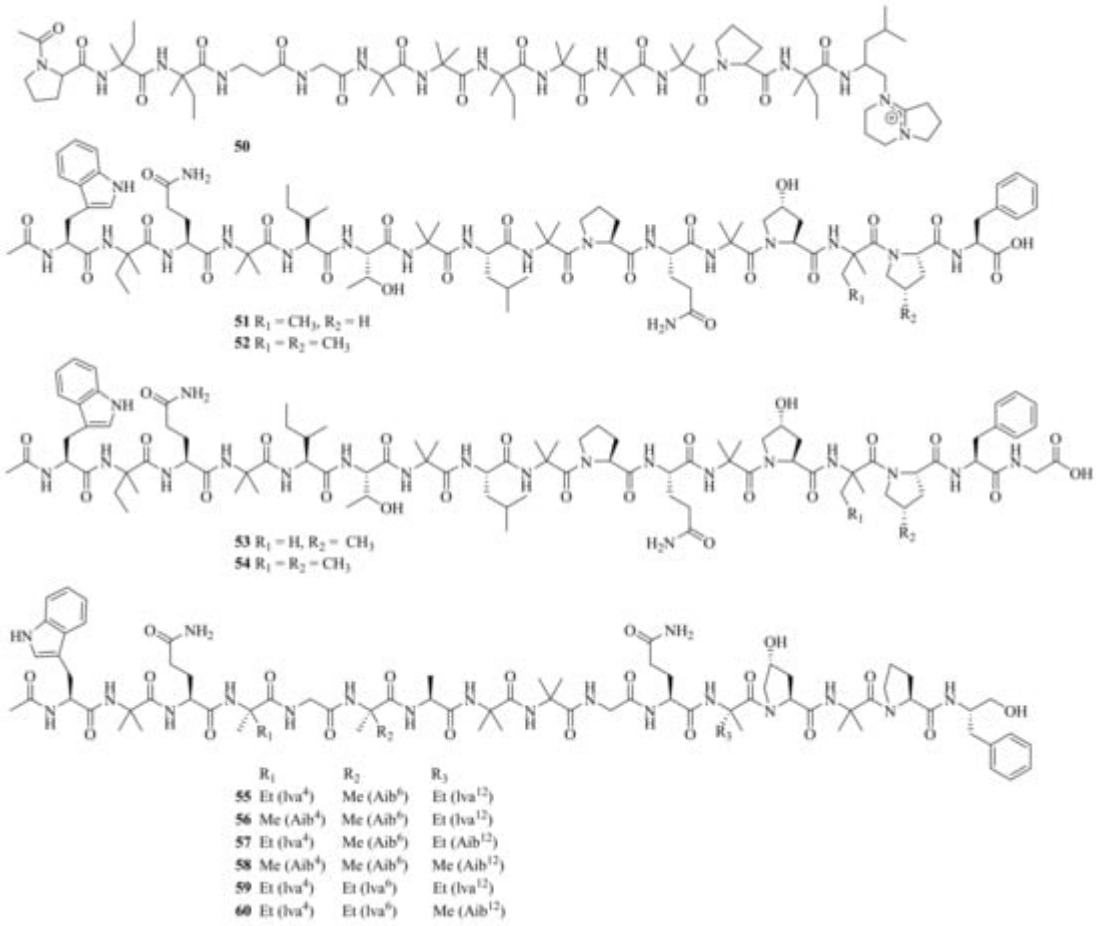

Figure 5. Chemical structures of linear peptides (**50**–**60**).

Six new 16-residue peptaibols, acremopeptaibols A–F (**55**–**60**), along with PF1171A, were isolated from the cultures of the sponge-associated fungus *Acremonium* sp. IMB18-086. Compounds **55** and **59** showed significant antibacterial activity against *S. aureus*, MRSA, *B. subtilis*, and *C. albicans*, with MIC values ranging from 16 to 64 μM [13]

Six new linear pentadecapeptides, emerimicins V–X (**61**–**66**) (**Figure 6**), were obtained from the soil-derived fungus *A. tubakii* MT053262. Emerimicins V (**61**) and VI (**62**) displayed strong toxicity toward Zebrafish embryos. In addition, emerimicin V showed certain activity against *Enterococcus faecalis*, MRSA, and vancomycin-resistant *Enterococcus faecium* with MIC values of 64, 32, and 64 μg/mL, respectively [19].

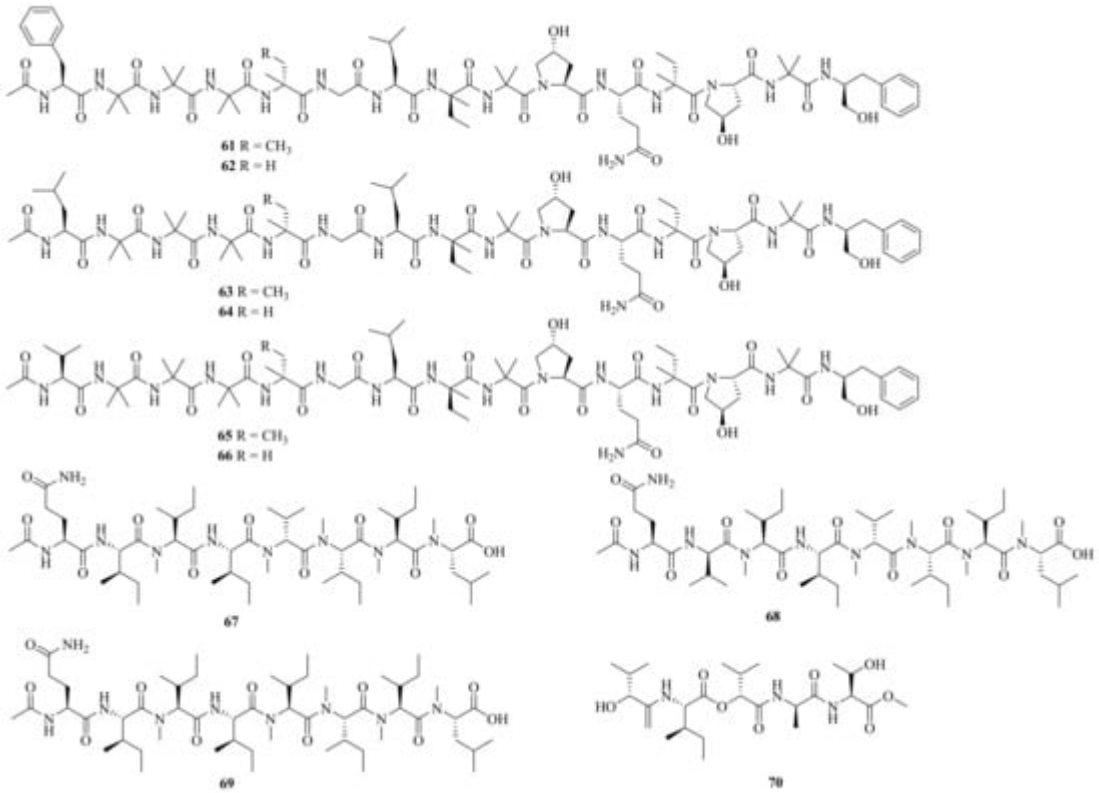

Figure 6. Chemical structures of linear peptides (**61**–**70**).

Four new peptides, acrepeptins A–D (**67**–**70**), and three known analogs, destruxin B, guangomide A, and guangomide B, were obtained from a marine algicolous fungus *Acremonium* sp. NTU492. Acrepeptins A (**67**) and B (**68**) exhibited significant inhibitory activity on NO production in LPS-activated microglia BV-2 cells, with IC_{50} values of 12.0 ± 2.3 and 10.6 ± 4.0 mM, respectively [20].

2.2. Cyclic Peptides

Four new hydroxamate-containing cyclopeptides, acremonpeptides A–D (**71**–**74**), together with a known one, Al (III)-acremonpeptide D, were obtained from the marine fungus *A. persicinum* SCSIO 115. Compounds **71**, **72**, and Al (III)-acremonpeptide D exhibited moderate antiviral activity against HSV-1 with EC_{50} values of 16, 8.7, and 14 μM, respectively [21] (**Figure 7**). Meanwhile, a new cyclic depsipeptide, acremonamide (**75**), was isolated from a marine-derived fungus *Acremonium* sp. strain CNQ-049 [22].

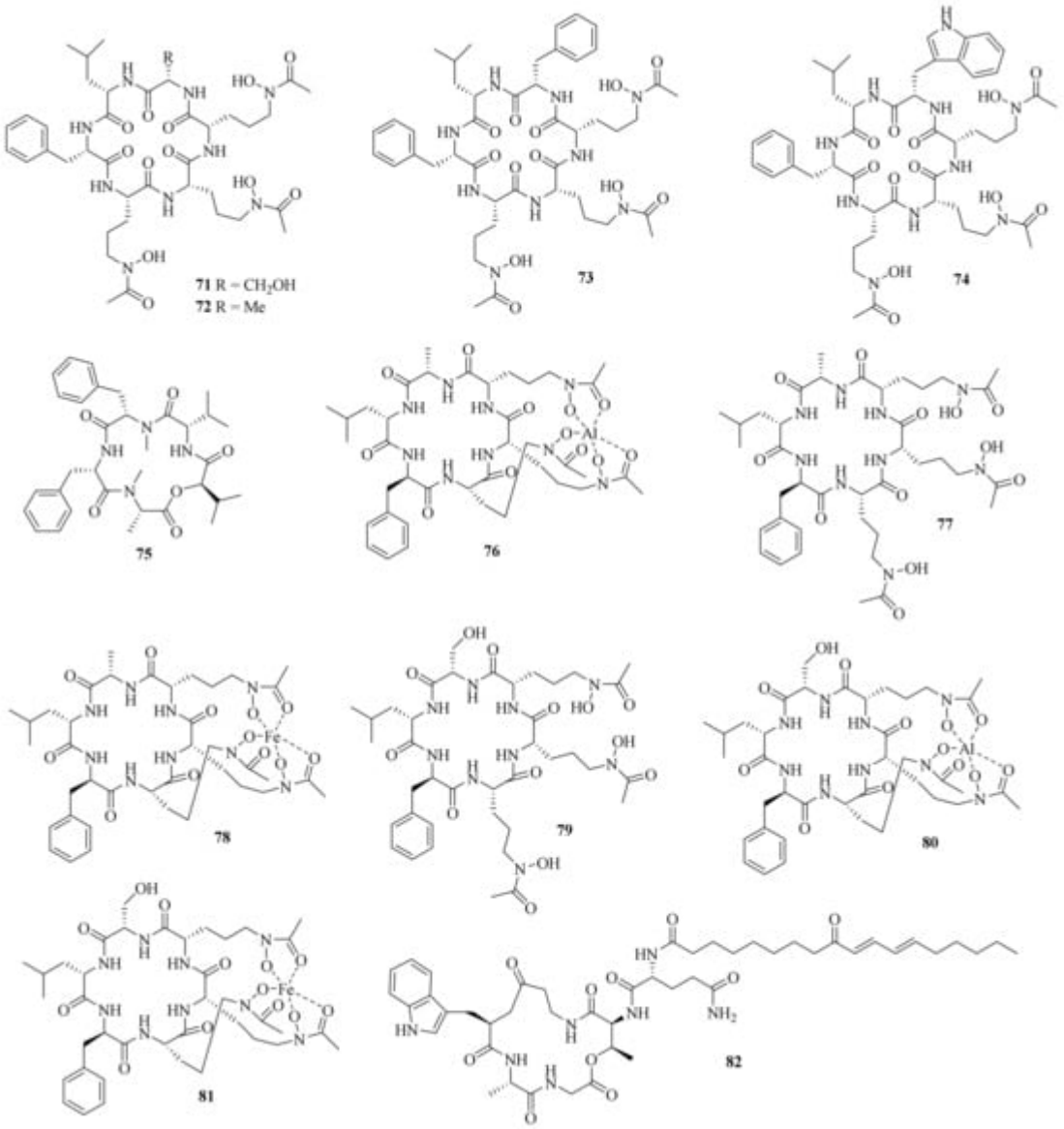

Figure 7. Chemical structures of cyclic peptides (**71**–**82**).

Six new hydroxamate siderophore cyclohexapeptides, Al (III)-acremonpeptide E (**76**), acremonpeptide E (**77**), Fe (III)-acremonpeptide E (**78**), acremonpeptide F (**79**), Al (III)-acremonpeptide F (**80**), and Fe (III)-acremonpeptide F (**81**), and one new cyclic pentapeptolide, aselacin D (**82**), together with a known compound, aselacin C, were isolated from the sponge-derived fungus *A. persicinum* F10. Compounds **76** and **80** showed pronounced antifungal activity against *Aspergillus fumigatus* and *A. niger* with a shared MIC value of 1 μg/mL, and both showed no cytotoxicity against human embryonic lung fibroblasts (MRC-5) at a concentration of 30 μM [23].

3. Polyketides

A total of 60 polyketides have been reported from the genus *Acremonium* within the period, including 23 new compounds and 18 bioactive compounds.

One new dibenzoquinone, 2,7-dihydroxy-3,6,9-trimethyl-9*H*-xanthene-1,4,5,8-tetraone (**83**) (**Figure 8**), and a known analog, 3,3′,6,6′-tetrahydroxy-4,4′-dimethyl-1,1′-bi-*p*-benzoquinon, were obtained from the fungus *A. cavaraeanum* CA022 [24]. Meanwhile, a chemical examination of marine sponge *Mycale* sp. derived fungus *A. persicinum* KUFA 1007 led to the isolation of one new compound, acremine T (**84**) [14].

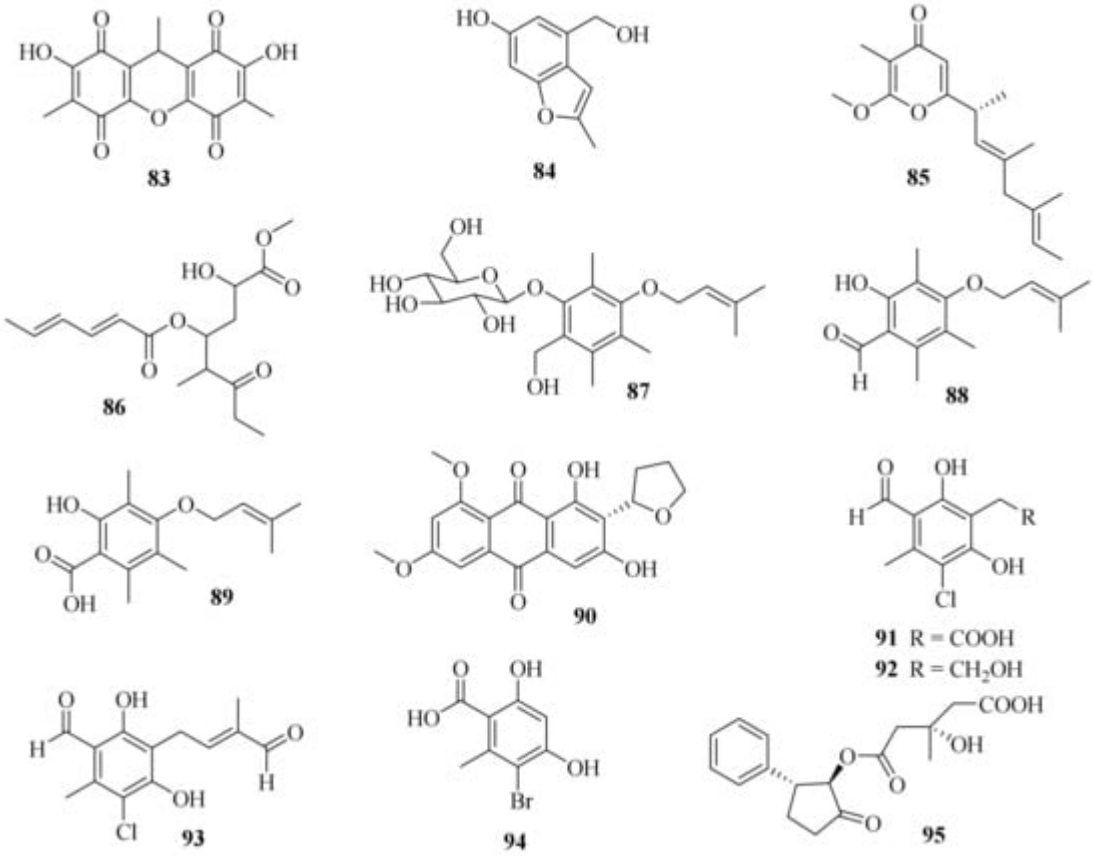

Figure 8. Chemical structures of polyketides (**83**–**95**).

A chemical investigation on the endophytic fungus *A. citrinum* SS-g13 yielded a new γ-pyrone derivative, acrepyrone A (**85**), and three known sorbicillinoids, trichodimerol, dihydrotrichodimerol, and tetrahydrotrichodimerol [25]. A chemical investigation of the endophytic fungus *A. citrinum* MMF4 derived from the root of the mangrove plant *Kandelia obovate* resulted in the isolation of one new compound, triacremoniate (**86**), along with a known compound, acrepyrone A. Compound **86** had significant inhibitory effects on the proliferation of HeLa cells, with an IC_{50} value of 30.5 ± 1.99 μM [26].

Three new zinniol analogues, pleoniols A–C (**87**–**89**), along with a known compound were isolated from a mixed fermentation of two endophytic fungi, *Pleosporales* sp. F46 and *A. pilosum* F47, both of which originated from the pedicel of the medicinal plant *Mahonia fortune* [27]. Four dimethylated anthraquinone derivatives, including one new compound, 6,8-di-*O*-methylbipolarin (**90**), and three known compounds, aversin, 6,8-di-*O*-methylaverufin, and 6,8-di-*O*-methylnidurufin, were obtained from the marine-derived fungus *A. vitellinum* MH726097. Compound **90** showed the strongest insecticidal activity against the third instar larvae of *Helicoverpa armigera*, with a LC_{50} value of 0.72 mg/mL [28].

Three new chlorinated orsellinic aldehyde derivatives, orsaldechlorins A–C (**91**–**93**), and one new natural brominated orsellinic acid, 5-bromo-2,4-dihydroxy-6-methylbenzoic acid (**94**), along with ten known biosynthetically related derivatives were further characterized from the Beibu Gulf coral-associated fungus *A. sclerotigenum* GXIMD 02501. Most of them inhibited LPS-induced NF-κB activation in RAW 264.7 cells at a concentration of 20 μM. Notably, compounds **91** and **92** showed inhibitions of RANKL-induced osteoclast differentiation in bone marrow macrophages without cytotoxicity [29].

One new compound, fusidione (**95**), along with a known one, microperfuranone, were isolated from the sea-water-derived fungus *A. fusidioides* RZ01. Fusidione displayed inhibitory activity against HL-60 cells with an IC_{50} value of 44.9 μM [30].

A new benzoyl compound, 1-(2′-benzoyl-3,4-dihydroxy-1′-methoxycyclobut-2′-enyl)-3,4,5-trihydroxy-2-methylnona-2,6-dien-1-one (**96**) (**Figure 9**), was obtained from the endophytic fungus *Acremonium* sp. of *Garcinia griffithii* [31]. One new polyketide, acrefurcatone A (**97**), was isolated from the deep-sea cold-seep sediment-derived fungus *A. furcatum* CS-280, which showed strong activity against *Pseudomonas aeruginosa* with an MIC value of 8 μg/mL [12].

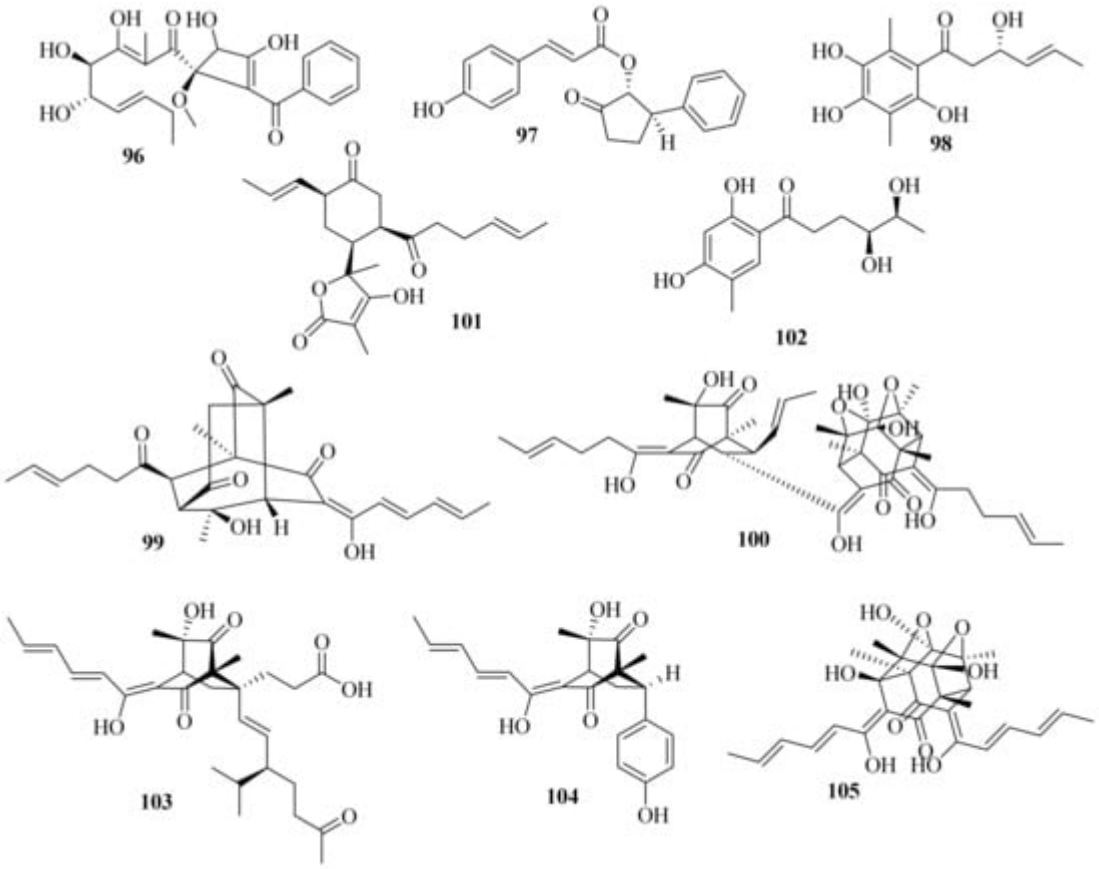

Figure 9. Chemical structures of polyketides (**96**–**105**).

A chemical investigation of marine sediment-derived fungus *Acremonium* sp. resulted in the isolation of two new compounds, 3(*S*)-hydroxy-1-(2,4,5-trihydroxy-3,6dimethylphenyl)-hex-4*E*-en-1-one (**98**) and acremonilactone (**99**),

along with eight known compounds. Among them, (2*E*,4*E*)-1-(2,6dihydroxy-3,5-dimethyl-phenyl) hexa-2,4-dien-1-one, sorbicillin, and tetrahydrotrichodimerol showed inhibitory activity against *S. aureus*, with a shared MIC value of 128 μg/mL. In addition, compounds **98** and trichodimerol showed 2,2-diphenyl-1-trinitrophenylhydrazine (DPPH) free radical scavenging activity with inhibition rates of 96.50% and 84.95% at a concentration of 0.5 mg/mL, respectively [32].

A chemical investigation of the terrestrial plant *Fructus mori* derived *A. citrinum* SS-g13 produced three new sorbicillinoids, trisorbicillinone E (**100**), acremosorbicillinoids A and B (**101** and **102**), and one new natural product, 2*S*,3*S*-acetyl-*β*-methyltryptophan, along with eight known sorbicillinoids. Among them, dihydrobisvertinolone showed significant cholesterol efflux-enhancing activity [33].

Moreover, three new sorbicillinoid derivatives, acresorbicillinols A–C (**103**–**105**), along with five known compounds were obtained from the marine-derived fungus *A. chrysogenum* C10. Compounds **104** and **105** displayed moderate activity against *S. aureus* and *Cryptococcus neoformans* with IC_{50} values of 86.93 ± 1.72 and 69.06 ± 10.50 μM, respectively. Moreover, compound **105** demonstrated strong DPPH free radical scavenging activity, with the IC_{50} value ranging from 11.53 ± 1.53 to 60.29 ± 6.28 μM in 24 h [34]. A chemical investigation of the deep-sea-derived *A. alternatum* provided two known bisorbicillinoids, tetrahydrotrichodimerol and dihydrotrichodimerol [35].

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