Natural sesquiterpenoids

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This review covers the isolation, structural determination, synthesis and chemical and microbiological transformations of natural sesquiterpenoids. 421 references are cited.

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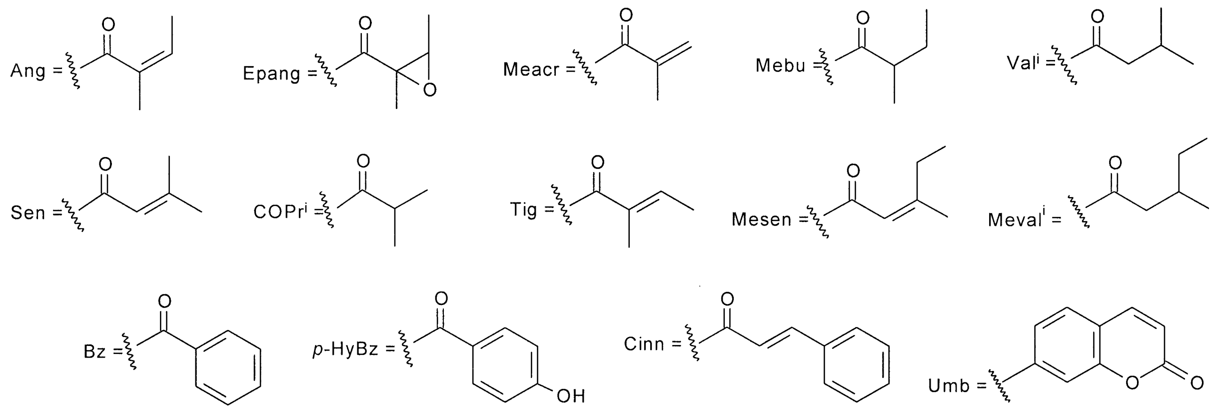
Braulio M. Fraga was born in Tenerife (1944) and received his PhD in Chemistry at the University of La Laguna (1970), where he lectured in Organic Chemistry for several years. In 1971 he was honoured with the Young Researcher Award of the Spanish Royal Society of Chemistry. He obtained a permanent position in the Spanish Council for Scientific Research as Tenured Scientist in 1972, being later appointed Research Scientist (1986) and Research Professor (1987). He was director of the Institute of Natural Products (Tenerife) from 1988–1991, and has been the representative of the Spanish Council for Scientific Research in the Canary Islands since 1991. He had previously been appointed Professor of Organic Chemistry at the University of Valencia (1981). His research interests range from the chemistry to the biotransformation of natural products, especially in the terpenoid field. He has authored two hundred scientific publications.

1 Introduction

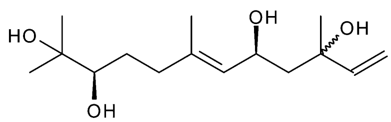
Sesquiterpenes have been used as chemosystematic markers in the subtribe Hypochaeridinae (Lactuceae tribe) of the Asteraceae family.[1] The chemical constituents in *Achillea*[2] and *Inula*[3] species have been reviewed. These genera are very rich in sesquiterpenes. Infrared spectroscopy has been utilised as a tool for chemotaxonomic studies within the *Achillea millefolium* group.[4] The chemical and biogenetic relationships among *Aristolochia* species from southeastern Brazil have been studied.[5] Twenty four sesquiterpene lactones with different skeleta have been investigated as IL-8 expression inhibitors in HeLa cells.[6] It has been shown that two pockets in the active site of maize sesquiterpene synthase (TPS4) are responsible for 14 different sesquiterpenes produced by this enzyme.[7]

2 Farnesane

The biological activity, source organism and country of origin of the linear furano- and pyrrolo-terpenoids have been reviewed.[8] Amaranthanolidols A–D **1–4** and amarantholidosides I–VII **5–11** are new nerolidol derivatives, which have been obtained from the weed *Amaranthus retroflexus*.[9,10] Five new sesquiterpene lactones **12–16**, and the known antheindurolides A and B, have been found in an extract from the aerial part of *Anthemis arvensis*.[11] In this



work the structure originally proposed of antheindurolide B has been revised to **17**. Four novel sesquiterpene peroxides **18–21** have been obtained from a Formosan soft coral of the *Sinularia* genus.[12]



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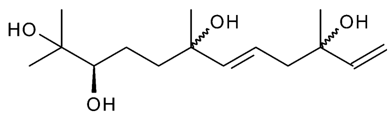

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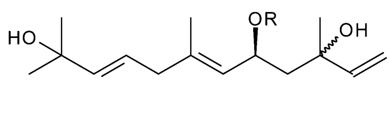
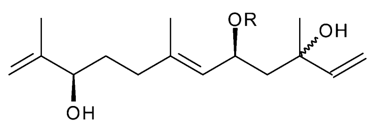


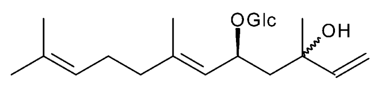
3 R = H
5 R = Glc



4 R = H
6 R = Glc

The biosynthesis of *trans*-(*S*)-nerolidol in fruits of *Fragaria* x *ananassa* (strawberry) has been studied using deuterated compounds. This work showed that this sesquiterpene is exclusively synthesised *via* the cytosolic mevalonic acid pathway.[13] It has been shown that an *Arabidopsis thaliana* methyl transferase is capable of methylating farnesoic acid.[14] 12-Fluorofarnesylphosphonophosphate has been synthesised as a potential inhibitor of sesquiterpene cyclases.[15] In an interesting review, C. D. Poulter has considered farnesyl diphosphate synthase to be "a paradigm for understanding structure and function relationships in *E*-polyprenyl diphosphate synthases".[16]

The first total synthesis of 6-oxodendrolasinolide has been accomplished.[17] Caulerpenyne, taxifolial A, dihydrorhipocephalin and furocaulerpin have been synthesised for the first time in racemic- and enantio-enriched forms. The *in vitro* inhibition of the polymerisation of microtubules by these sesquiterpenes has also been investigated.[18] 2-Methyl-1,4-benzoquinone has been used as



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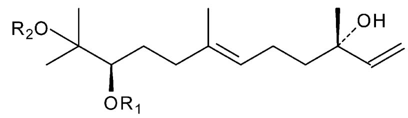
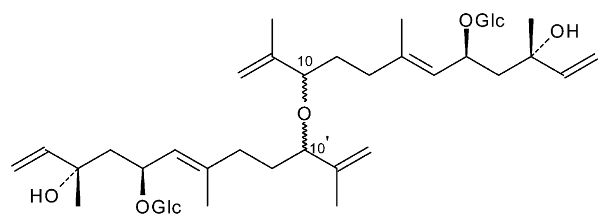
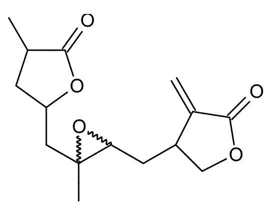


8 R_1 = Glc R_2 = H
9 R_1 = H R_2 = Glc



10 (10*R*,10'*R*) or (10*S*,10'*S*)
11 (10*S*,10'*S*) or (10*R*,10'*R*)



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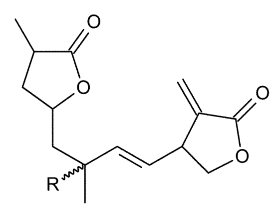
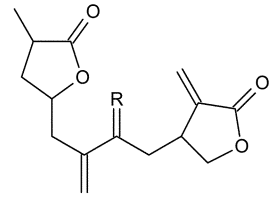
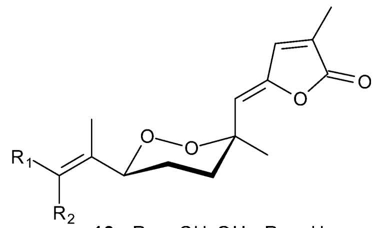


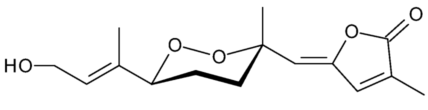
13 R = OOH
14 R = OH



15 R = OOH,H
16 R = OH,H
17 R = O



18 $R_1 = CH_2OH$ $R_2 = H$
19 $R_1 = H$ $R_2 = CH_2OH$



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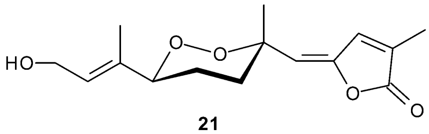

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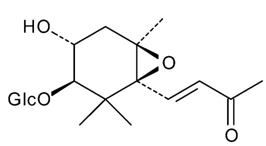
starting material in the synthesis of two triprenylated toluquinones and two toluhydroquinone derivatives, which had been isolated from a marine-derived *Penicillium* fungus.[19] A study of the superacid cyclisation of certain aliphatic sesquiterpene derivatives in ionic liquids has been carried out.[20] Several sesquiterpenoids, isolated from *Ferula fukanensis*, have been shown to inhibit nitric oxide production by a murine macrophage-like cell line.[21]

3 Monocyclofarnesane

An α-ionone derivative **22** has been found in the aerial parts of *Salsola tetrandra*.[22] Bridelionosides A–F **23–28** and debilosides A–C **29–31** are megastigmane glucosides, which have been isolated from *Bridelia glauca*[23] and *Equisetum debile*,[24] respectively. Other compounds of this type, macarangiosides A–D **32–35**, obtained from the leaves of *Macaranga tanarius*, showed potent DPPH radical-scavenging activity.[25] A study of the components of *Youngia japonica*[26] led to the isolation of the glucoside **36**. An inamoside derivative, named cuneatoside, has been found in an extract from the leaves and branches of *Erythroxylum cuneatum*,[27] whilst vignoside **37**, a glucoside that inhibits the growth of human stomach cancer, has been obtained from *Vigna angularis* (adzuki bean).[28] Several glycosides of blumenol C, 13-hydroxy-blumenol C and 13-*nor*-5-carboxy-blumenol C accumulate when



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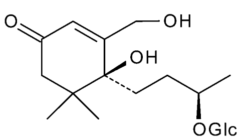

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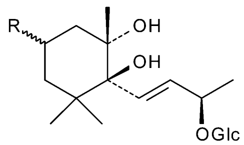
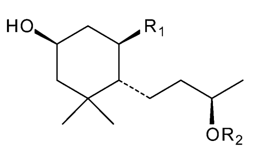
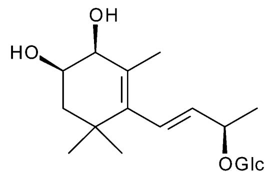


24 R = α-OH
25 R = β-OH



26 $R_1 = CH_2O$ H R_2 = Glc
27 $R_1 = CO_2Glc$ R_2 = H



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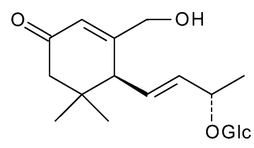

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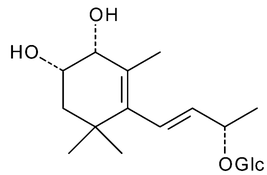

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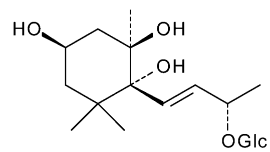

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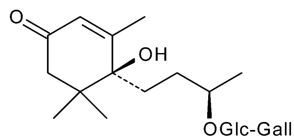

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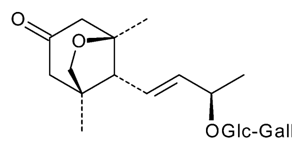
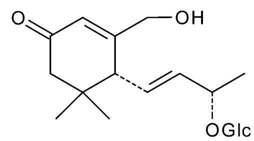


33
34 Dihydro



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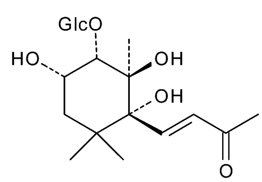

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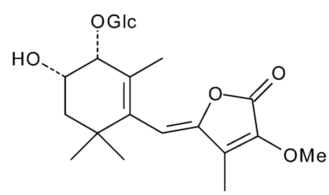

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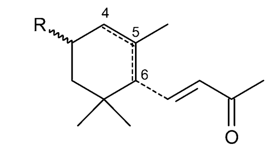
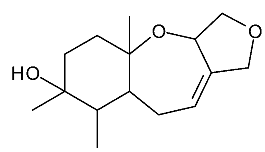
roots of *Ornithogalum umbellatum* are colonised by an arbuscular mycorrhizal fungus.[29] A carotenoid dioxygenase from melon, that cleaves carotenoids generating ionone derivatives, has been characterised and cloned.[30]

Two new megastigmane derivatives, **38** and **39**, have been obtained from the red alga *Gymnogongrus flabelliformis*.[31] Another red alga *Laurencia luzonensis* contains five novel sesquiterpenes **40–44**, two of which **40** and **41** possess a new rearranged snyderane skeleton.[32]

Racemic α-ionone has been used as starting material for syntheses of the enantiomeric forms of α- and γ-damascone[33] and of 3,4-didehydroionone stereoisomers.[34] Enantioselective syntheses of (+)-ricciocarpin A[35] and striatenic acid[36] have been achieved. A total synthesis of the *trinor*-cyclonerolidane sesquiterpene (−)-chokol A has been reported.[37]



38 Δ^{4} R = β-OCO-CH_2-C_6H_5
39 $\Delta^{5(6)}$ R = α-OCO-CH_2-C_6H_5



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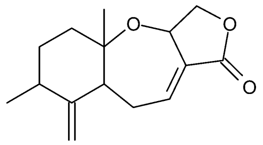

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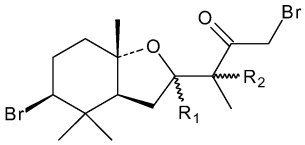
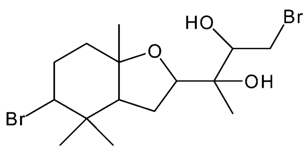


42 R_1 = β-H R_2 = α-OH
43 R_1 = α-H R_2 = β-OH



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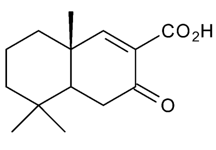
A 9-*cis*-epoxycarotenoid dioxygenase inhibitor has been used in the elucidation of the mechanisms of action of abscisic acid.[38] A concise and enantioselective synthesis of this plant hormone, and of a new analogue, has been reported.[39] (+)-AHI4 is a new inhibitor of ABA 8′-hydrolase, which has been synthesised with an α-axial hydroxyl group instead of the geminal methyls at C-6′ characteristic of (+)-AHI1, (1′*S*,2′*S*)–(+)-6-*nor*-2′3′-dihydro-4′-deoxy-ABA.[40] Bicyclic analogues of abscisic acid have been shown to have enhanced biological activity in plants.[41]

4 Bicyclofarnesane

Nebularic acids A **45**, nebularic acid B **46**, nebularilactone A **47** and nebularilactone B **48** are four new drimane sesquiterpenes, which have been found in the fungus *Lepista nebularis*.[42] The fruiting bodies of *Stereum ostrea* contain the new sesquiterpene methoxylaricinolic acid.[43]



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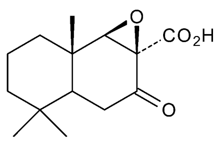

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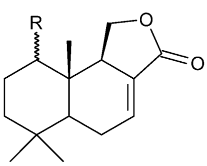
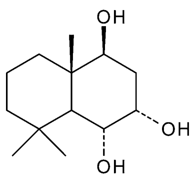


47 R = β-OH
48 R = α-OH

The novel *dinor*-sesquiterpene japonicumin D **49** has been obtained from *Lycopodium japonicum*.[44] A phytochemical analysis of the tree *Pleodendron costaricense*[45] led to the isolation of the drimane derivative parritadial **50**. The fungicidal sesquiterpene paxidal **51** has been found in a foliage extract from a New Zealand shrub, *Pseudowintera axillaris*, using a bioactivity-directed separation. This sesquiterpene possesses fungicidal activity against the plant pathogen *Phytophthora infestans*.[46] The structures of cinnamolid-3β-ol hemihydrate and 3β-hydroxycinnamolide acetate, two components of *Warburgia ugandensis*, have been determined by X-ray analysis.[47] Other compounds from this plant have been evaluated as inhibitors of 12(*S*)-HETE and of leukotriene metabolism.[48] The dimeric and trimeric drimane sesquiterpenes cinnafragrins A–C **52–54** have been found in an extract from the medicinal plant *Cinnamosma fragrans*.[49] In this work, the structure of capsicodendrin has been revised to a mixture of the C-12′ epimers **55**.



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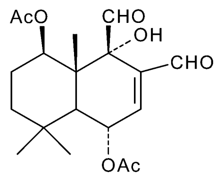

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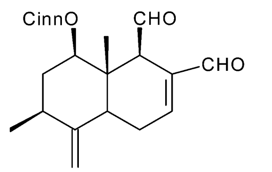

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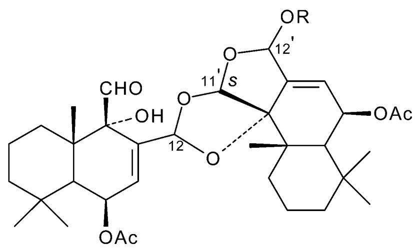
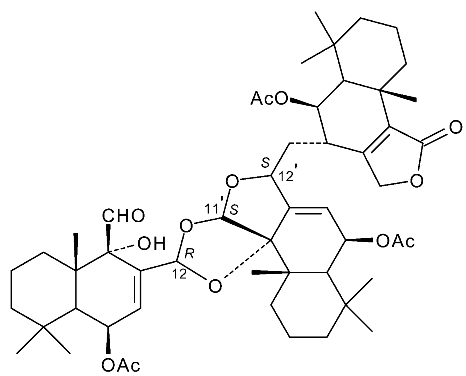


52 C-12 (*S*), C-12' (*S*) R = H
53 C-12 (*R*), C-12' (*S*) R = Me
55 C-12 (*R*), C-12' (*R* and *S*) R = H



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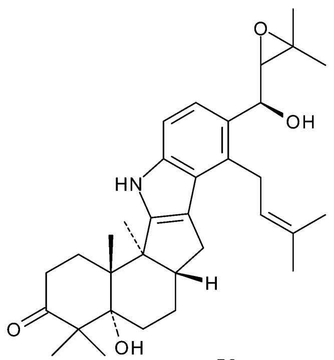
Sespendole **56** is a novel bioactive indolosesquiterpene, which has been isolated from the culture broth of the fungus *Pseudobotrytis terrestris* strain FKA-25.[50–52] The structures of dasyscyphins



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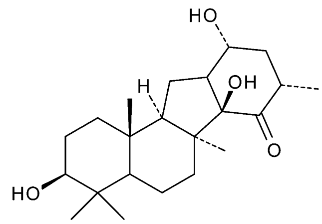

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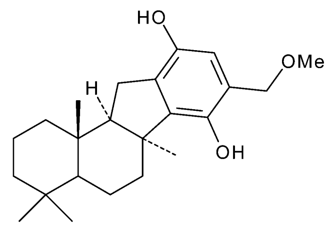

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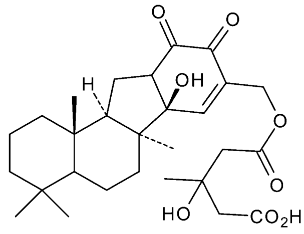

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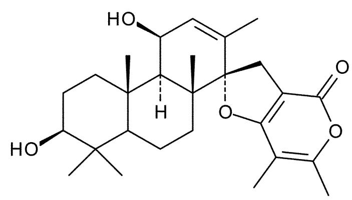

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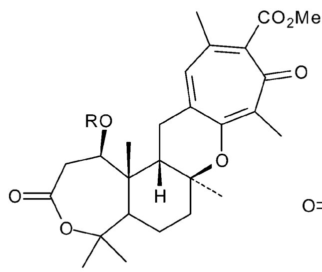
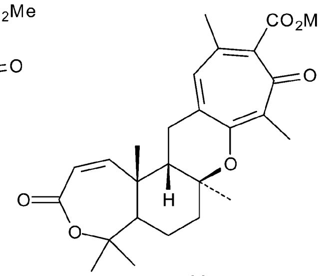
A–C and niveulone have been determined as **57–59** and **60**, respectively. These meroterpenoids have been obtained from the ascomycete *Dasyscyphus niveus*.[53–55] Other compounds of this type with cytotoxic properties, tropolactones A–D **61–64**, have been isolated from a marine derived fungus *Aspergillus* sp., which was obtained from an unidentified sponge, collected at Manele Bay in Hawaii.[56] Chromatography of an extract from an unidentified species of the *Dysidea* genus afforded the bioactive sesquiterpene **65**, which has been named avinosol.[57] Three new species of this genus have been characterised by their sesquiterpenoid content. Thus, sesquiterpene-hydroquinones are the main components of *Dysidea reformensis* and *Dysidea cachui*, while the furanosesquiterpene dendrolansin was obtained from *Dysidea uriae*.[58] The reactivity and biological activity of the marine sesquiterpene hydroquinone avarol, and related sesquiterpenes, have been reviewed.[59]

The biotransformation of (−)-Ambrox® by cell suspension cultures of kiwifruit, *Actinidia deliciosa*, has been investigated.[60] Stereoselective syntheses of 9-*epi*-Ambrox®,[61] and Superambrox®,[62] and racemic syntheses of hyphodermin B[63] and pallescensin A,[64] have been reported. Drimane dienes with functional groups at C-6 have been synthesised by photodegradation of larixol derivatives.[65] (±)-Wiedendiol B and a siphonodictyal B derivative have been prepared using a new approach to the synthesis of sesquiterpene arenes.[66] The diterpene (+)-manool has been used as starting material for the preparation of the sesquiterpene hydroquinone (+)-*ent*-chromazonarol.[67] Another diterpene, sclareol oxide, has been employed in the synthesis of *ent*-thallusin,[68] whilst a bioinspired strategy has been used in the synthesis of hongoquercin A and rhododaurichromanic acid A.[69] A highly diastereoselective formal synthesis of puupehedione and 8-*epi*-puupehedione has been described.[70] A short sequence of reactions has been used in the preparation of bicyclic sesquiterpene units linked to aromatic structures, which has



61 R = H
62 R = Ac



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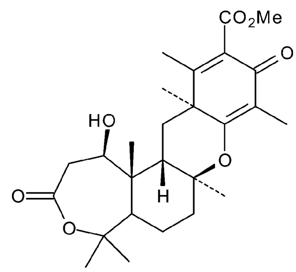

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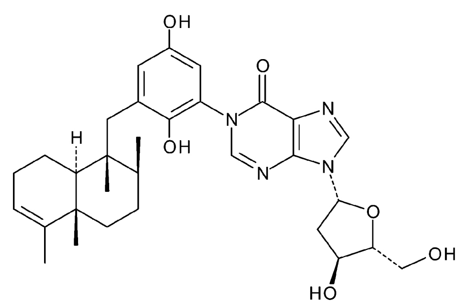
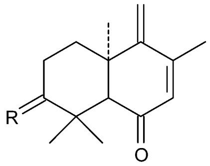


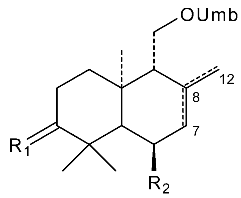
65

permitted formal syntheses of zonarol, zonarone, puupehedione and umbrosone.[71] A rapid method for the construction of the A, B and C rings of terreulactone A has been reported.[72]

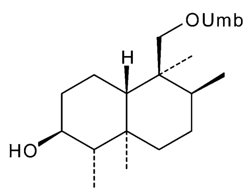
The drimane sesquiterpenes fetidone A **66** and fetidone B **67** have been found in an extract from assafetida. This gum resin also contains known sesquiterpene coumarin ethers, one of which, (8-acetoxy)-5-hydroxyumbelliprenin, was showed to be a potent NF-κB inhibitor,[73] while conferone **68**, isolated from *Ferula schtschurowskiana*, enhanced the cytotoxicity of vinblastine in MDCK-MDR1 cells.[74] The sesquiterpene coumarin saradaferin **69** has been obtained from *Ferula asafoetida.*[75] Other new compounds of this type, isofeterin **70**, lehmannolol **71** and sinkianone **72**, have also found in extracts from the roots of *Ferula sinkiangensis* and *Ferula teterrima*.[76]



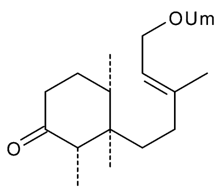
66 R = O
67 R = α-OH,H



68 Δ^{7} R_1 = O R_2 = H
69 $\Delta^{8(12)}$ R_1 = α-OH,H R_2 = H
70 $\Delta^{8(12)}$ R_1 = β-OH,H R_2 = OAc



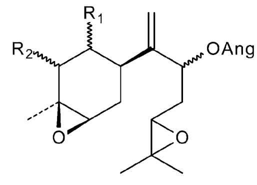
71



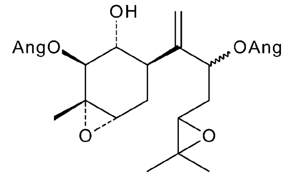
72

5 Bisabolane, sesquicarane, heliannane group, majapolane and parvifolane

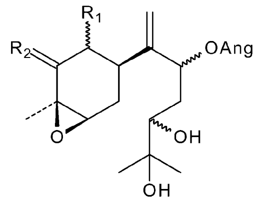
The male-produced sex pheromones from the Brazilian rice stalk stink bug, *Tibraca limbativentris*, has been characterised as isomers of 1′*S*-zingiberenol.[77] Eight new bisabolane sesquiterpenes **73–80** have been isolated from the roots of *Ligularia cymbulifera*.[78] Another compound of this type **81** has been found in an extract from *Achillea clavennae*.[79] The aerial parts of *Lippia dulcis*[80] contain six novel bisabolane sesquiterpenes, which have been named peroxylippidulcines A–C **82–84**, peroxyepilippidulcine B **85**, epilippidulcine B **86** and epilippidulcine C **87**. The bisabolane derivatives boivinianin A **88** and boivinianin B **89** have been identified as components of the stem bark of *Cipadessa boiviniana* (Meliaceae).[81] Another three new sesquiterpenes **90–92** have been isolated from the roots of *Leontopodium longifolium*.[82,83] Baccharisketone **93** is another new bisabolane derivative, which has been obtained from the leaves of *Baccharis dracunculifolia*.[84] The active principle of an extract from the roots of *Ostericum koreanum*[85] which affects adults of the *Dermatophagoides* genus, has been identified as the known sesquiterpene bisabolangelone **94**.



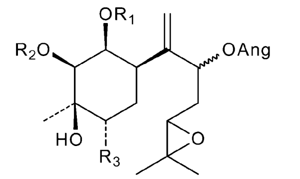
73 R_1 = α-OH R_2 = α-OAng
74 R_1 = R_2 = β-OAng
75 R_1 = β-OAng R_2 = H



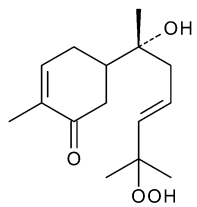
76



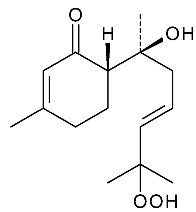
77 R_1 = β-OAng R_2 = β-OH,H
78 R_1 = α-OAng R_2 = O



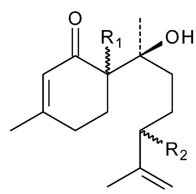
79 R_1 = H R_2 = Ang R_3 = OAng
80 R_1 = Ang R_2 = H R_3 = Cl



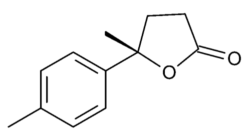
81



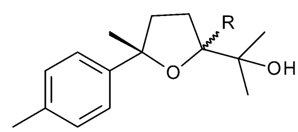
82



83 R_1 = β-H R_2 = β-OOH
84 R_1 = β-H R_2 = α-OOH
85 R_1 = α-H R_2 = β-OOH
86 R_1 = α-H R_2 = β-OH
87 R_1 = α-H R_2 = α-OH

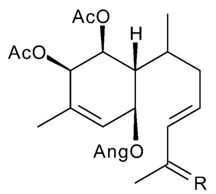


88

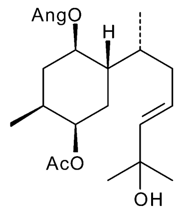


89 R = α-H and β-H

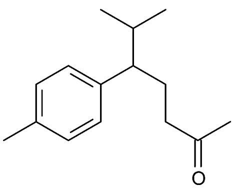
A species of sponge of the *Myrmekioderma* genus,[86] collected at the Vanuatu coast, contains the bisabolane sesquiterpenes **95–97**. Three other novel halogenated β-bisabolene derivatives **98–100** have been isolated from the red alga *Laurencia scoparia*,[87] whilst the aldingenins B–D **101–103** have been obtained from another species of this genus *Laurencia aldingensis*.[88] The sesquiterpenoids **104–107** have been found in an extract from the sea hare *Aplysia dactylomela*.[89] β-Bisabonol has been synthesised using a ring-closing olefin metathesis reaction in the key step.[90]



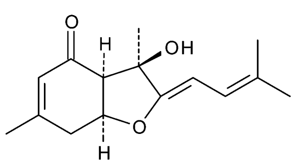
90 R = CH_2
91 R = Me,OH



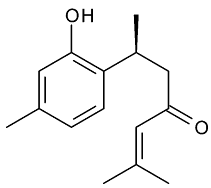
92



93



94



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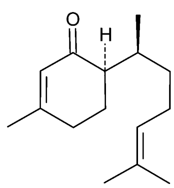

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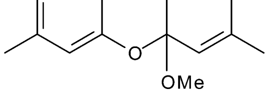

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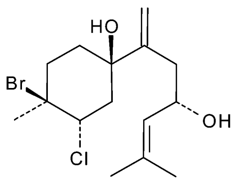

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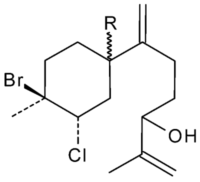
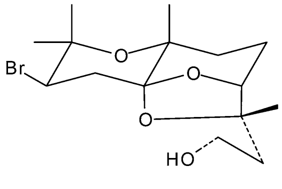


99 R = β-OH
100 R = α-OH



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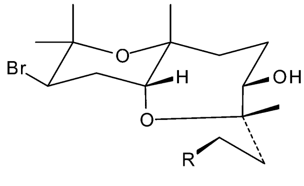
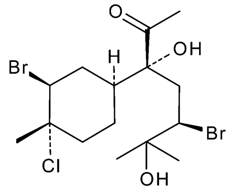


102 R = H
103 R = OAc

Heliespirone B **108** and heliespirone C **109** are two sesquiterpenes with two novel spiro heterocyclic skeleta, which have been isolated from *Helianthus annuus*.[91] The structures of heliannuols G and H, also obtained from this species, have been revised to **110** and **111**, respectively, as a consequence of enantiocontrolled total syntheses.[92] This plant also contains heliannuol C, whose synthesis has also been accomplished.[93] A total synthesis of sesquicarene **112** has been described.[94] This sesquiterpene had been isolated from *Schisandra chinensis*.[95] The absolute configurations of majapolene



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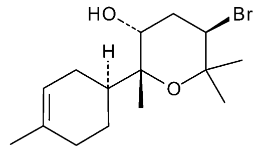

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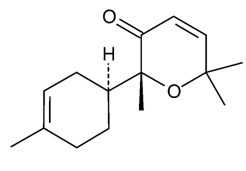

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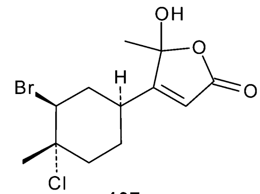
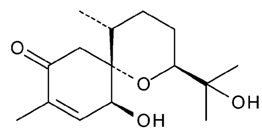


107

B **113** and acetylmajapolene B **114** have been determined by vibrational circular dichroism. These sesquiterpenes have been found in an extract from a *Laurencia* sp., collected in Malaysia.[96] The first enantioselective synthesis of (−)-parvifoline and (−)-curcuquinone has been reported.[97]



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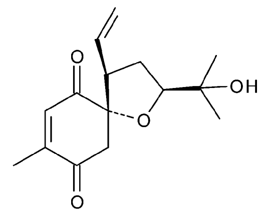

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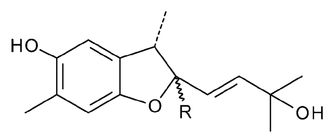
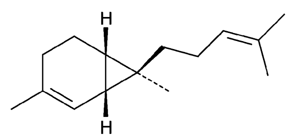


110 R = β-H
111 R = α-H



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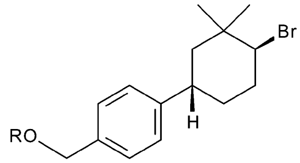
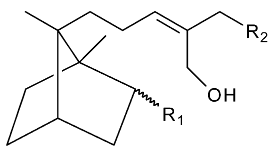


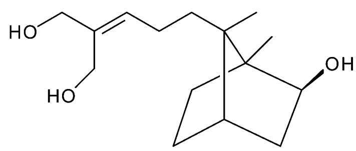
113 R = H
114 R = Ac

6 Sesquicamphane, cyclosesquicamphane, campherenane, fumagillane and petasitane

Syntheses of two (*E*)-*endo*-bergamoten-12-oic acid derivatives have been reported.[98] Three new campherenane, **115**, **116** and **119**, and three new santalane sesquiterpenes **121–123**, with bioactive properties, have been isolated from *Santalum album* of Indian origin.[99] In this work, the absolute structure of **117**, **118** and **120** has also been established. These compounds had been obtained from commercial sandalwood.[100] In this plant, *Santalum album*, a quantitative chemotaxonomic approach has been used to study the biosynthetic relationships between the sesquiterpenes isolated from the heartwood.[101] The effects of inhalation of East Indian



115 R_1 = α-OH R_2 = OH
116 R_1 = β-OH R_2 = OH
117 R_1 = α-OH R_2 = H
118 R_1 = β-OH R_2 = H



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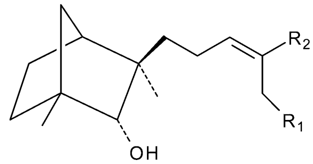
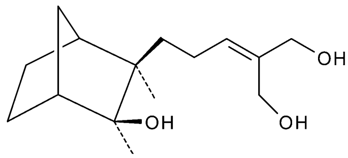
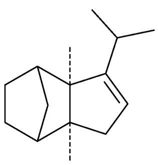


120 R_1 = OH R_2 = CH_3
121 R_1 = OH R_2 = CH_2OH
122 R_1 = H R_2 = CHO



123

sandalwood essential oils and its main compound, α-santalol, on human physiological parameters, have been measured in healthy volunteers.[102] Concise, enantio- and diastereo-selective total syntheses of fumagillol, RK-805, FR65814, ovalicin and 8-demethylovalicin have been accomplished.[103] The rare sesquiterpene hydrocarbons petasitene **124** and albene **125** have been identified as components of a root oil obtained from *Artemisia vulgaris* of Serbian origin.[104]



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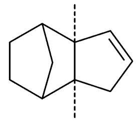
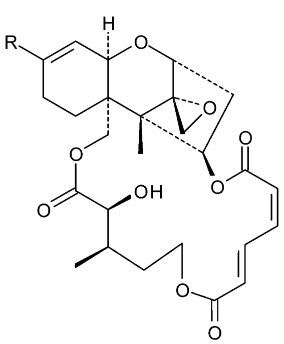


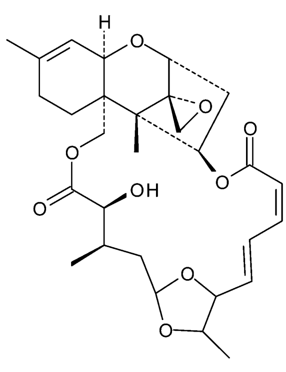
125

7 Trichothecane, cyclotrichothecane, cuparane, laurane, cyclolaurane and herbertane

Two highly cytotoxic compounds, 16-hydroxyverrucarin A **126** and verrucarin X **127**, have been isolated from fermentations of the fungus *Myrothecium roridum*.[105] Another three macrocyclic trichothecenes, 12′-hydroxyroridin E, roridin Q and 2′,3′-deoxyroritoxin D, have been obtained from two strains of the marine-derived fungus *Myrothecium roridum* TUF 98F42, whilst roridin R **128**, and the known roridin A, roridin H and isororidin E, have been found in *Myrothecium* sp TUF 02F6.[106] Other strains of *Myrothecium roridum*, IFB-E009 and IFB-E012, which were isolated as endophytic fungi from the Chinese medicinal plants

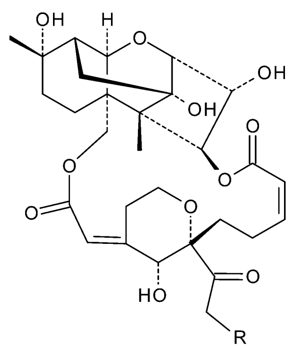


126 R = CH_2OH
127 R = CO_2H

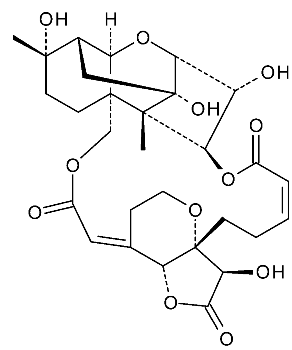


128

Trachelospermum jasminoides and *Artemisia annua*, respectively, contain the new cyclotrichothecane macrolides myrothecines A–C **129–131**. The absolute configuration of mytoxin B was also established in this study.[107] Another cyclotrichothecane derivative miophytocen C **132**, together with 3-hydroxyroridin E and 13′-acetyltrichoverrin B, were isolated from a saltwater culture of *Myrothecium verrucaria*, separated from a *Spongia* sp., collected in Hawaii.[108]



129 R = H
130 R = OH



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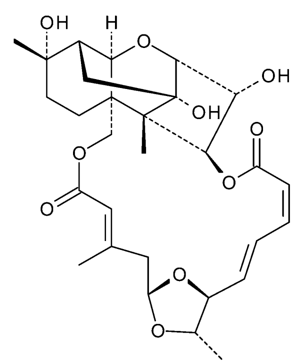
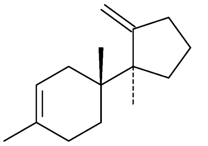


132

GC–MS techniques have been used in the analysis of trichothecene mycotoxins in commercial corn harvested in Brazil.[109] The synthesis of four mycotoxins of this type labelled with carbon-13 and their application as internal standards in stable isotope dilution assays for their determination in foods has been described.[110] The loss of toxicity of deoxynivalenol in extruded cereal-based products has been confirmed using different types of analysis.[111]

Two new pathways for the enzyme-catalysed formation of trichodiene **133** have been proposed, one of which, a proton-transfer pathway, appears to be much more energetically favourable than the hydride transfer pathway usually proposed.[112] The structures of two new bazzanene derivatives have been determined as **134** and **135**. These sesquiterpenes have been isolated from a New Zealand liverwort, *Frullania falciloba*.[113]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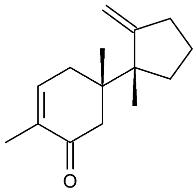
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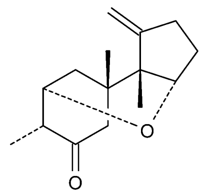
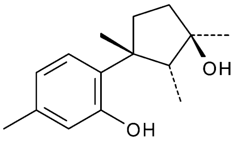

135

A new sesquiterpene, 3,7-dihydroxydihydrolaurene **136**, has been obtained from the red alga *Laurencia obtusa*, whilst a dimeric sesquiterpene of the cyclolaurane type **137** was isolated from *Laurencia microcladia*. These algae had been collected from the coastal rocks of Serifos in the Aegean Sea.[114] Another species of this genus, *Laurencia okamurai*,[115] contains a new laurane derivative, 3β-hydroxyaplysin **138**, and two novel rearranged sesquiterpenes, laurokamurene A **139** and laurokamurene B **140**. The structure of the laurokamurene A was identical to a metabolite which had been obtained from *Laurencia microcladia* (see our last review[116]). The structure assigned to HM-3, a sesquiterpene which had been isolated from the phytopathogenic fungus *Helicobasidium mompa*, has been found to be incorrect by total synthesis of the proposed structure.[117] In this work the first synthesis of HM-3 **141** and HM-4 **142** has also been carried out. An enantioselective synthesis of (−)-α-herbertenol has been achieved.[118] (±)-β-Cuparenone has been synthesised in a short and efficient procedure.[119] A. Srikrishna *et al.* have carried out racemic total syntheses of herbertenediol,[120] grimaldone, epigrimaldone, α-cuparenones[121] and lagopodin A,[122] and a racemic formal synthesis of cuparene and herbertene.[123]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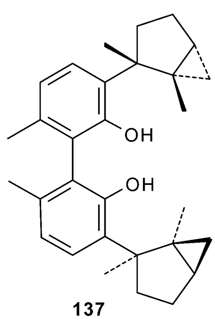
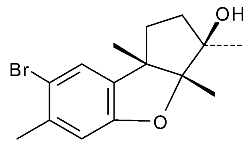

137

8 Chamigrane, perforane, perforetane and widdrane

The thalloid liverwort, *Reboulia hemisphaerica*, contains two new *ent*-β-chamigrane derivatives **143** and **144**. The biotransformation of the latter by the fungus *Aspergillus niger* has also been described in this work.[124] A gas chromatographic method has been developed to quantify the chamigrane sesquiterpene elatol, at the surface and within the thallus, of 70 specimens of the red alga *Laurencia obtusa*.[125] This species also contains the new sesquiterpenes perforenol B **145** and the perforetane derivative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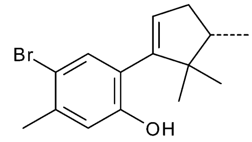
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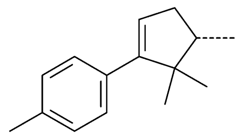
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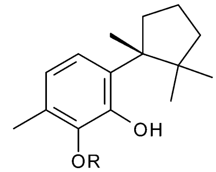
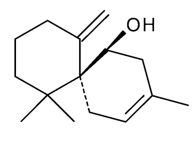

141 R = Ac
142 R = H

146.[114] Total syntheses of racemic α-chamigrene, β-chamigrene and laurencenone C have been described.[126]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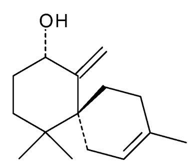
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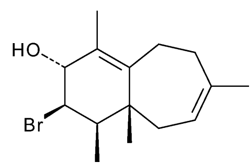
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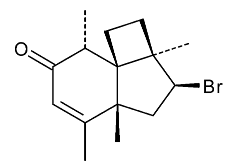
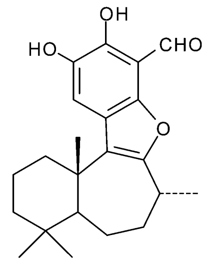

146

The structure elucidation and biomimetic synthesis of liphagal **147** has been carried out. This selective inhibitor of PI3 kinase has been obtained from the sponge *Aka coralliphaga*.[127] The antifungal activity of widdrol and its microbiological transformation by the fungi *Colletotrichum gloeosporioides* and *Botrytis cinerea* have been investigated.[128]

9 Carotane, acorane, cedrane, zizaane and anislactone group

A formal synthesis of (±)-α-cedrane and (±)-β-cedrane has been reported.[129] After 25 years of the alquene-arene *meta*-photocycloaddition reaction, which was developed in a synthesis of α-cedrene, a review has appeared on the application of this reaction to the synthesis of other natural products.[130] D. E. Cane *et al.* have described the molecular cloning and characterisation of a new sesquiterpene synthase from *Streptomyces coelicolor*. This enzyme catalyses the cyclisation of farnesyl diphosphate to a novel sesquiterpene, *epi*-isozizaene **148**, which has not been isolated as a natural product, but which had been chemically prepared by acid treatment of (+)-zizaene.[131]



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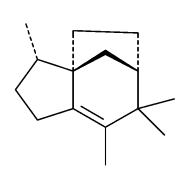
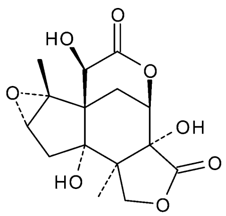


148

Three new secoprezizaane sesquiterpene lactones **149–151** have been obtained from *Illicium micranthum*.[132] Racemic syntheses of jiadifenin,[133] merrilactone A[134] and 11-*O*-debenzoyltashironin[135] have been accomplished.



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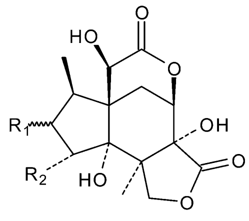
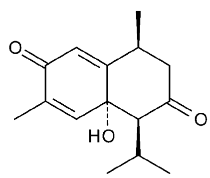


150 R_1 = β-OH R_2 = H
151 R_1 = α-OH R_2 = OH

10 Cadinane, cubebane, oplopane and helminthosporane

The new sesquiterpene **152** has been isolated from an aqueous extract of *Malva silvestris*.[136] A phytochemical study of *Commiphora myrrha*[137] led to the isolation of six new cadinane derivatives, which have been named myrracadinols A–C **153–155** and myrracalamenes A–C **156–158**. The sesquiterpene **159** and the norsesquiterpene eupatorone **160** have been obtained from the flowers of *Eupatorium adenoforum*.[138] Other authors have isolated another novel sesquiterpenoid **161** from the leaves of this species.[139] Two new cadinane derivatives **162** and **163** have been found in an extract from the bark of *Jatropha neopauciflora*,[140] while the aerial parts of *Pseudoelephantopus spicatus*[141] has been shown to contain a new lactone, named spicatocanolide A **164**. Bioassay-guided studies of an ethanol extract from *Swartzia polyphylla* afforded T-cadinol as the metabolite responsible for the larvicidal and antimycobacterial activity of this Peruvian plant.[142] Pubinernoid C **165** is a new cadinane sesquiterpene, which has been obtained from *Schisandra pubescens*.[143] Three novel dehydroabietane diterpenes sugikorojins G–I **166–168**, which incorporate a cadinane sesquiterpene in their structure, have been isolated from *Cryptomeria japonica*,[144,145] whilst a diterpene-sesquiterpene ether **169** has been obtained from *Calocedrus macrolepis*, and named calocedimer B.[146] Chromatography of an extract from *Phomopsis cassiae*, an endophytic fungus associated with *Cassia spectabilis*, afforded five new cadinane derivatives **170–174**.[147] A patent describing the cytotoxic activity of the dimers parviflorene H and parviflorene I has appeared.[148] Another cadinane sesquiterpene, amentotaxone **175**, has been isolated from *Amentotaxus formosana*.[149] The cloning and functional characterisation of a *cis*-muuroladiene synthase from *Mentha piperita* has been described.[150]



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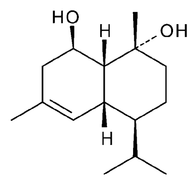

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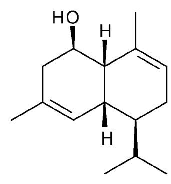

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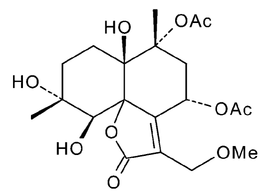

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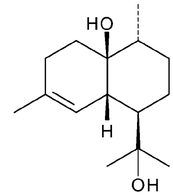

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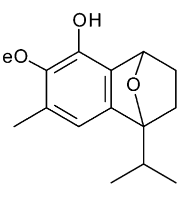

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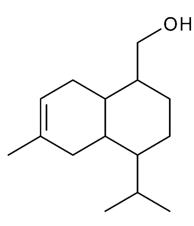

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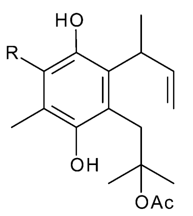
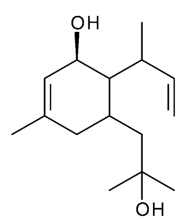


154 R = H
155 R = OH



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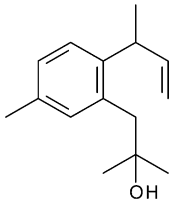

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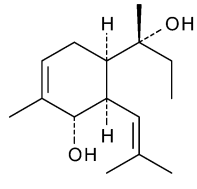

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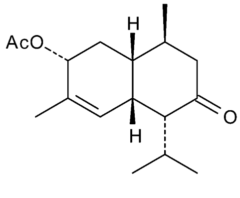

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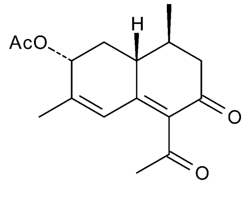

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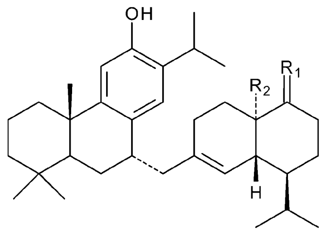
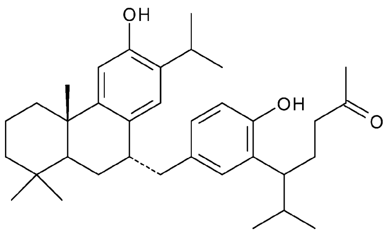


166 R_1 = α-OH,Me R_2 = H
167 R_1 = β-OH,Me R_2 = OH



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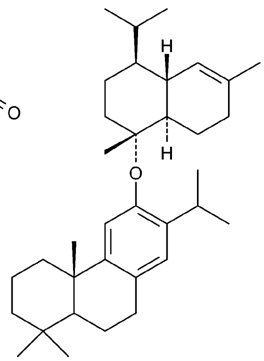

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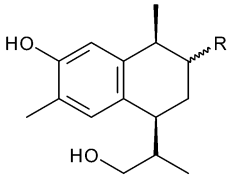
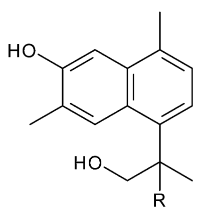
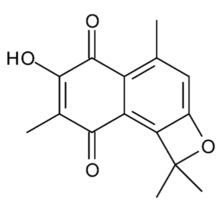


170 R = β-OH
171 R = α-OH
172 R = H

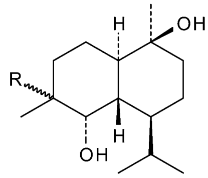


173 R = H
174 R = OH

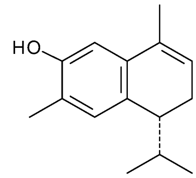


175

The structures of 4α,5α,10β-trihydroxycadinane **176** and its C-4 epimer **177** had been assigned to two sesquiterpenes, which had been isolated from *Jasonia candicans*[151] and *Taiwania cryptomerioides*,[152] respectively. Total syntheses of these compounds have now been carried out, indicating that both sesquiterpenes are identical, and possess the second of these structures.[153] The first total synthesis of xenitorin B and xenitorin C has been achieved, allowing the determination of their absolute configuration.[154] Syntheses of 7,8-dihydroxycalamenene and mansonone C have been achieved.[155] An alternative procedure for the synthesis of mansonone F and of a biflorin precursor has been described.[156] The structure of isocalamendiol has been determined by X-ray analysis.[157] This technique has also been applied to the determination of the absolute configuration of (−)-3-hydroxy-α-calacorene **178**.[158]

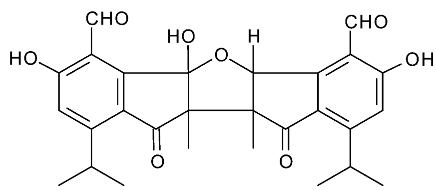


176 R = α-OH
177 R = β-OH



178

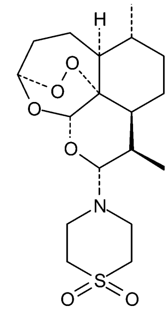
6-Methoxygossypol and 6,6′-dimethoxygossypol have been obtained from the seed and root bark from a cotton variety growing on St. Vincent island.[159] The ratios of (+)-gossypol and (−)-gossypol in leaves, stems and roots from selected accessions of *Gossypium hirsutum* have been determined.[160] The effects of these compounds, and of their racemic forms, on the survival and development of *Helicoverpa zea* larvae have been evaluated.[161] The peroxidative coupling of hemigossypol to (+)-gossypol and (−)-gossypol in extracts of cottonseed has been investigated.[162] Aquatidial **179** is a new bis-norsesquiterpene, which has been isolated from the roots of *Pachira aquatica*. This compound is probably formed by the dimerisation of a cadinane derivative, isohemygossypolone, which was also obtained from this plant.[163]



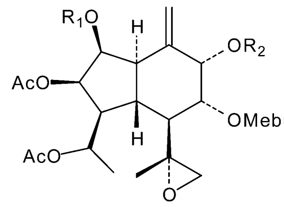
179

The biosynthesis of artemisinin and its regulation in *Artemisia annua* have been reviewed.[164] The mechanism and stereochemistry of the cyclisation of farnesyl diphosphate into amorpha-4,11-diene by amorpha-4,11-diene synthase, a key enzyme in artemisinin biosynthesis, have been studied.[165,166] A cDNA clone, encoding a cytochrome P450, catalyses the oxidation of amorpha-4,11-diene, artemisinic alcohol and artemisinic aldehyde, which are intermediates in the biosynthesis of artemisinin.[167] The effects of overexpression of the endogenous farnesyl diphosphate synthase on the artemisinin content in *Artemisia annua* have been investigated.[168] Density functional theory calculations have been applied to a theoretical study of the reductive decomposition of this antimalarial sesquiterpene.[169] The production of artemisinic acid in an engineered strain of the yeast *Saccharomyces cereviasae* has been accomplished by J. D. Keasling *et al.* These authors first manipulated the FPP biosynthetic pathway to increase the production of this intermediate by the yeast, then introduced a gene, which transforms FPP to amorpha-4,11-diene, and finally introduced a cytochrome P450 that converts this diene into artemisinic acid.[170] The fungus *Streptomyces griseus* has been used in the biotransformation of a novel ketone-derivative of artemisinin.[171]

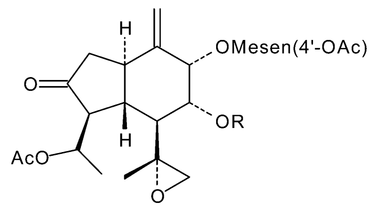
A paper describing a comparative assessment of the established and emerging technologies for the extraction of artemisinin has appeared.[172] The accumulation of this antimalarial sesquiterpene during the development and senescence of the leaves of *Artemisia annua* has been studied.[173] A simple route to access new 10-fluoro-artemisinin derivatives has been reported.[174] The reaction



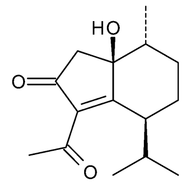
180



181 R_1 = Mebu R_2 = Mesen(4'-OAc)
182 R_1 = H R_2 = Mesen
183 R_1 = Mebu R_2 = Mesen(4'-OH)
184 R_1 = Mebu R_2 = Mesen



185 R = Mebu
186 R = Meval^{i}



187

mechanism of 12 antimalarial artemisinin derivatives with two competitive pathways has been studied by means of quantum chemical calculations.[175] A new highly active antimalarial drug of the artemisinin class, named artemisone **180**, has been described,[176] whilst new orally active derivatives of artemisinin, with high efficacy against multidrug-resistant malaria in mice, have been developed.[177] The anti-angiogenic activity of several deoxoartemisinin derivatives has been reported,[178] whilst the antiviral effect of artemisinin against the bovine viral diarrhoea virus has been investigated.[179] Artemisinin and extracts of the aerial parts of *Artemisia annua* have been evaluated as anti-insect products.[180]

A stereoselective synthesis of (−)-cubebol has been accomplished.[181] Other authors have reported the synthesis of this sesquiterpene and of the hydrocarbon (−)-α-cubebene.[94] New oplopane sesquiterpenes **181–186** have been isolated from *Ligularia narynensis*.[182,183] Another compound of this type **187** has been found in an extract from the brown alga *Dictyopteris divaricata*.[184] New helminthosporal analogues with plant growth regulatory properties have been synthesised.[185]

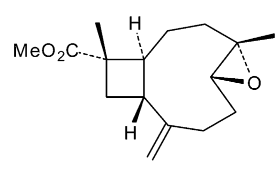
11 Himachalane, longifolane and longipinane

Four known himachalane sesquiterpenes have been identified in volatiles collected from the eggplant flea beetle, *Epitrix fuscula*.[186] An enantioselective total synthesis of (+)-β-himachalene has been achieved.[187] The conformations of longifolene[188] and of a longipinene derivative[189] have been studied by NMR and molecular mechanics calculations. The microbiological transformation of (+)-cycloisolongifol-5β-ol by the fungus *Cunninghamella elegans* has been investigated.[190]

12 Caryophyllane, modhephane, isocomane, silphinane, presilphiperfolane, silphiperfolane, pethybrane, botryane and quadrane

The caryophyllane derivative **188** has been found in extracts from the Formosan soft coral *Sinularia gibberosa*.[191] (−)-β-Caryophyllene has been identified as a gender-specific sesquiterpene emitted by the multicoloured Asian lady beetle, *Harmonia axyridis*.[192] Known caryophyllane sesquiterpenes have been isolated from *Aframomun arundinaceum*,[193] *Pulicaria prostrata*[194] and *Zingiber nimmonii*.[195] The biotransformation of (−)-caryophyllene oxide by cell suspension cultures of *Catharanthus roseus*[196] and by several species of fungi[197] have been described. The transannular cyclisation of epoxycaryophyllenes catalysed by Ti^{III} has been investigated.[198] The absolute configuration of birkenal has been determined by chemical correlation with a caryophyllene derivative.[199]

Three new sesquiterpenes 6-hydroxypunctaporonin A **189**, 6-hydroxypunctaporonin B **190** and 6-hydroxypunctaporonin E **191** have been obtained from cultures of the fungicolous fungus *Pestalotiopsis disseminata*.[200]



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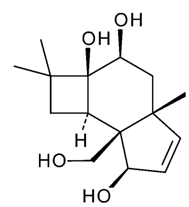

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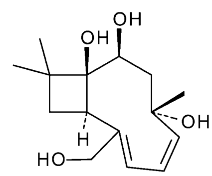

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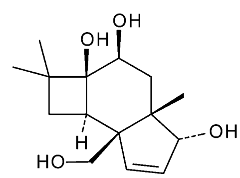
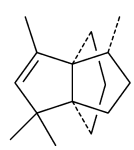


191

The acid-catalysed rearrangement of (−)-modhephene **192** and (−)-isocomene **193** to a (−)-triquinane **194** has been studied.[201] The silphinene derivative **195** has been found in cultures of an isolate of the fungus *Penicillium griseofulvum*. This metabolite is a probable precursor of other sesquiterpenes, which were also obtained from this species and named penifulvins A–E **196–200**,



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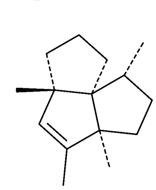

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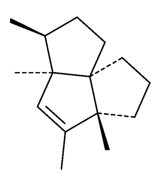

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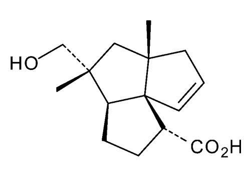
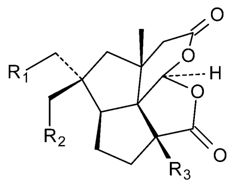
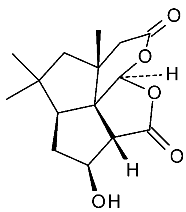


195

and which possess a new carbon skeleton.[202,203] The uncommon sesquiterpene hydrocarbons silphiperfol-5-ene **201**, modhephene **192**, pethybrene **202**, presilphiperfol-7-ene **203**, silphin-1-ene **204** and isocomene have been identified as components of a root oil obtained from *Artemisia vulgaris*.[104]



196 $R_1 = R_2 = R_3 = H$
197 $R_1 = OH$ $R_2 = R_3 = H$
198 $R_1 = R_3 = H$ $R_2 = OH$
199 $R_1 = R_2 = H$ $R_3 = OH$



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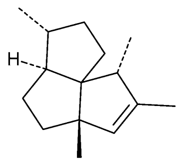

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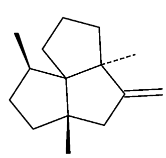

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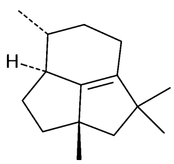

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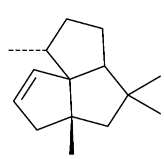
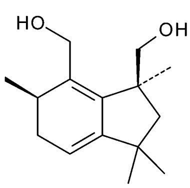


204

The role of botrydienediol **205** in the biodegradation of the sesquiterpenoid phytotoxin botrydial **206** by *Botrytis cinerea* has been investigated.[204] The absolute configurations of quadrone **207**, suberosenone, suberosanone and suberosenol A acetate have been determined *via* density functional theory calculations of optical rotation, electronic circular dichroism and vibrational circular dichroism.[205] A patent describing the use of a sesquiterpene ketone, isolated from *Subergonia suberosa*, as an anticancer drug has appeared.[206]



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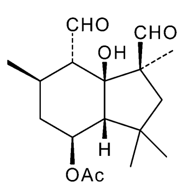

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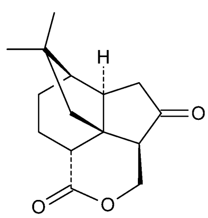
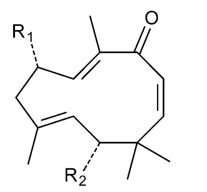


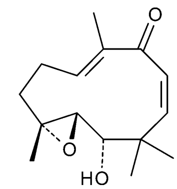
207

13 Humulane, hirsutane, lactarane, capnellane, protoilludane, illudane, illudalane, marasmane, pentalenane, tremulane and africanane

The humulane sesquiterpenes mitissimols A–C **208–210**, and a mixture of mitissimol A oleate and mitissimol B linoleate, have been isolated from the fruiting bodies of *Lactarius mitissimus*.[207] Another compound of this type, 10ξ-acetoxyhumula-3,7-diene, has been obtained from the roots of *Casearia multinervosa*.[208] The Sharpless asymmetric epoxidation of (+)-zerumbol has been studied.[209] The structures of hirsutenols D–F have determined as **211–213**. These sesquiterpenes have been found in an extract of the culture broth of *Stereum hirsutum*.[210] Chemoenzymatic syntheses of (+)-hirsutic acid and (−)-complicatic acid have been achieved.[211] Rapraesentins D–F **214–216** and 1,2-dehydrolactarolide A **217** are new plant growth regulators, which have been isolated from the fungi *Lactarius repraesentaneus*[212] and *Lactarius vellereus*,[213] respectively. A group of lactarene derivatives have been tested as antiviral, cytotoxic, antiproliferative and immunotropic agents.[214]



208 $R_1 = H$ $R_2 = OH$
210 $R_1 = R_2 = OH$



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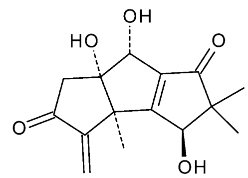

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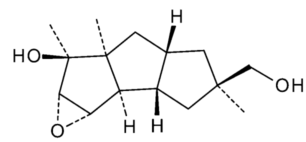

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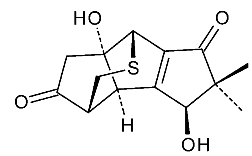

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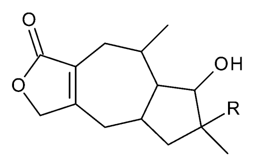
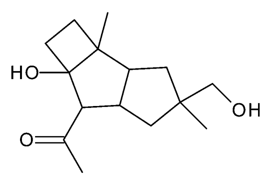


214 $R = CH_2OH$
215 $R = CO_2H$



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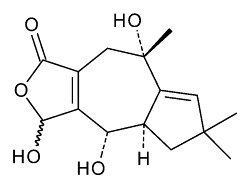
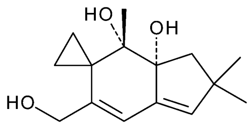
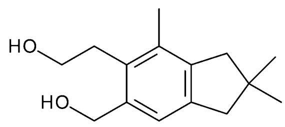


217

The basidiomycete *Ripartites metrodii* contains three new sesquiterpenes, riparols A–C **218–220** with an illudane, illudalane and protoilludane skeleton, respectively.[215] The russujaponols A–F **221–226** are six novel illudane sesquiterpenes, which have been obtained from the fruiting body of *Russula japonica*.[216] Chromatography of an extract from the cultured mycelia of the fungus *Echinodontium japonicum*[217] afforded two illudalane sesquiterpenes, which have been named echinolactone C **227** and echinolactone D **228**. An approach to the synthesis of (−)-coprinolone, a protoilludane derivative, has been reported.[218] The synthesis and biological evaluation of alcyopterosin A **229**



218



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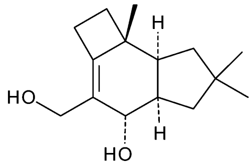

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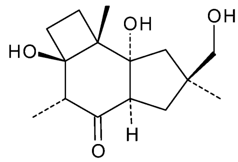

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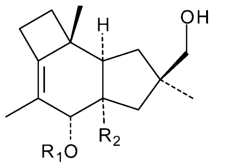
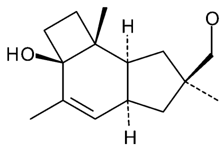


222 R_1 = Ac R_2 = OH
223 R_1 = R_2 = H



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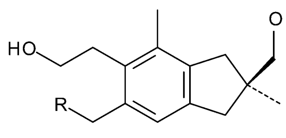
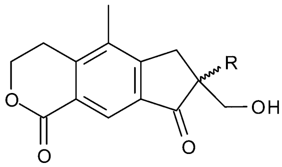
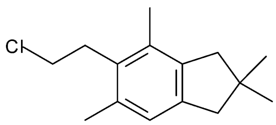


225 R = OH
226 R = H



227 R = β-Me
228 R = α-Me

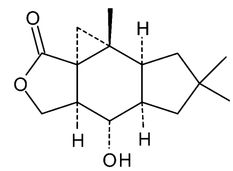


229

and other illudalane derivatives, as anticancer agents, has been described.[219]

The synthesis of the tricyclic core of the 5α-capnellenols has been reported.[220] A total synthesis of the sesquiterpene dichomitol has revealed that the structure assigned to this compound was erroneous.[221] Two new marasmane sesquiterpenes, **230** and **231**, have been isolated from *Russula foetens*.[222]

The existence in *Streptomyces avermitilis* of a gene cluster for the biosynthesis of pentalenolactone **232** has been reported.[223] The molecular cloning and assignment of the biochemical functions of PtlH and PtlI, a non-heme iron dioxygenase[224] and a cytochrome P450,[225] respectively, have been described. The first, in this fungus, converts 1-deoxypentalenic acid to 11β-hydroxy-1-deoxypentalenic acid, whilst the second transforms pentalenene to pentalen-13-al. Quantum chemical computation studies have been carried out with the aim of ascertaining possible polycyclisation pathways of the farnesyl cation into pentalenene.[226] An enantiospecific approach towards the synthesis of pentalenolactone has been described.[227]



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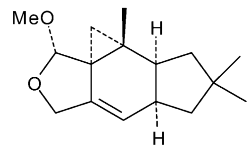

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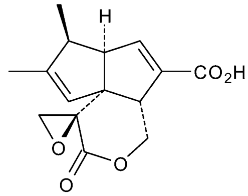
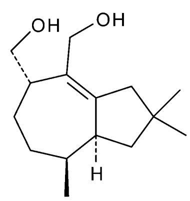


232

Enantioselective syntheses of tremulenediol A **233** and tremulenolide A **234** have been accomplished.[228] The new africanane alcohol **235** has been isolated from the essential oil of *Lippia integrifolia*.[229]



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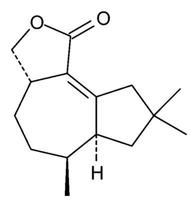

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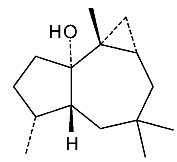


235

14 Germacrane

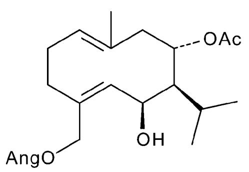
The new sesquiterpene **236** has been found in an extract from the roots of *Thapsia nitida*.[230] The known sesquiterpenes germacrane D and kunzeanol have been isolated from the essential oil of *Brickellia veronicaefolia*[231] and from the leaf surface of *Solanum tuberosum*,[232] respectively. Other known germacrane derivatives have been obtained from the marine octocoral *Muricea austera*.[233] The cloning, expression, purification and characterisation of recombinant germacrane A and germacrene D synthases from *Artemisia annua*[234] and *Zingiber officinale*,[235] respectively, have been described. The pheromone periplanone C has been synthesised as its racemate.[236]

The new germacrane lactones which have been isolated from natural sources during 2006 are listed in Table 1. The structures **237–259** represent the new germacranolides, while the structures **260–262** have been assigned to the new heliangolides and **263–264** to the melampolides. No new *cis*,*cis*-germacranolides have been obtained this year.

The structure **257** given to vernolide C[247] proved to be the same as that reported for vernchinilide A (see our last review[116]). The full paper on the isolation of the lactones eupanilonides A and D from *Eupatorium lindleyanum* has appeared.[251] Known germacranolides have been obtained from *Camchaya calcarea*[252] and *Ichthiothere terminalis.*[253] It has been shown that the sesquiterpene lactone parthenolide induces apoptosis of human acute myelogenous leukaemia stem cells,[254] and of B-chronic limphocytic

Table 1 Sources of germacrane lactones

Source	Germacranolides	Ref.
Achillea asplenifolia	**237–242**	237
Achillea collina	**243**	238
Dimerostemma brasilianum	**244**	239
Eupatorium chinense	**245**	240
Eupatorium kiirunense	**246**	241
Glechoma longituba	**247**	242
Helianthus annuus	**248–251**	243
Inula britannica	**252**	244
Mulgedium tataricum	**253**, **254**	245
Pseudoelephantopus spicatus	**255**	141
Sarcandra glabra	**256**	246
Vernonia cinerea	**257**, **258**, See text	247
Vernonia triflosculosa	**259**	248
Heliangolides		
Eupatorium kiirunense	**260**, **261**	241
Eupatorium lindleyanum	**262**	249
Melampolides		
Siegesbeckia orientalis	**263**, **264**	250



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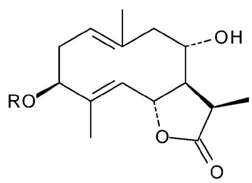
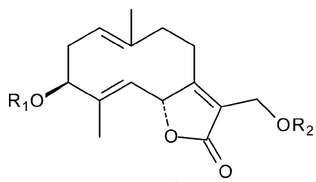
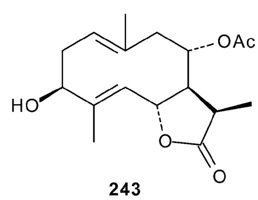


237 R = Val^i
238 R = Mebu



239 R$_1$ = Val^i R$_2$ = Ac
240 R$_1$ = Mebu R$_2$ = Ac
241 R$_1$ = Val^i R$_2$ = H
242 R$_1$ = Mebu R$_2$ = H



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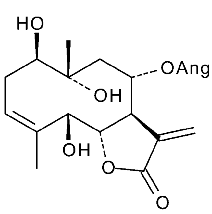
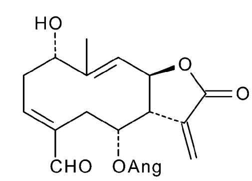
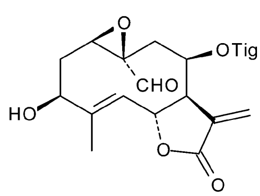


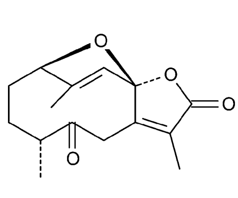
244



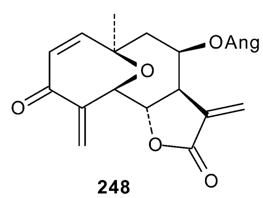
245 Eupatochinilide I



246 Eupakirunsin I



247



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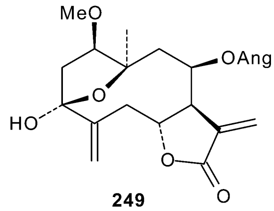
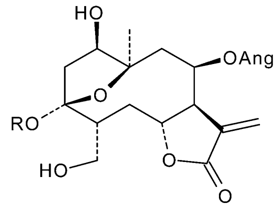
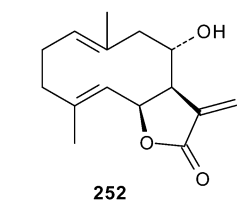


249



250 R = Me
251 R = Et



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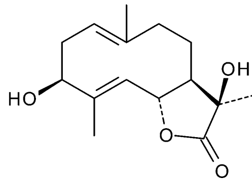

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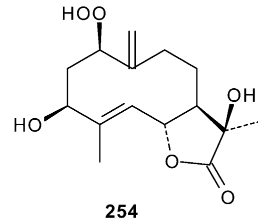

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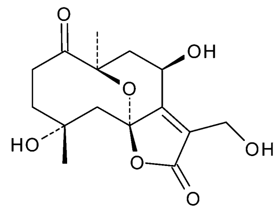
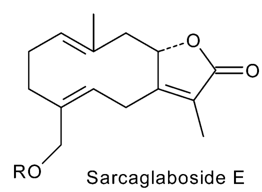
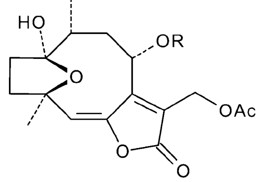


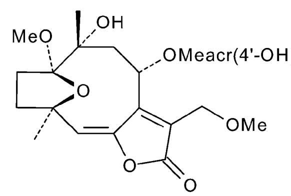
255 Spicatolide C



Sarcaglaboside E
256 R = Glc-(6'-1'')-Api



Vernolides C and D
257 R = COPr^i(2'-OH,3'-Cl)
258 R = Tig(4'-OH)



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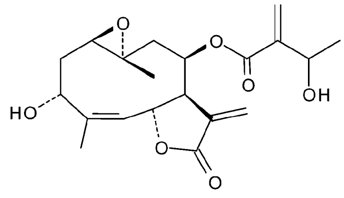
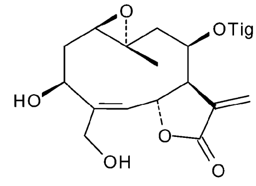
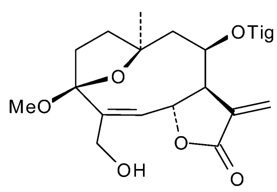


260 Eupakirunsin F



261 Eupakirunsin G

leukaemia cells.[255] The synthesis and antiviral activity of a series of sesquiterpene lactones, structurally analogous to parthenolide, have been reported.[256] The lactone cnicin has been shown to be a potent and irreversible inhibitor of the antibacterial target enzyme MurA.[257] The anionic and photochemical behaviour of the hallerin derivative **265** have been studied.[258] Several 13-amino costunolide derivatives have been synthesised and evaluated as anticancer agents.[259]



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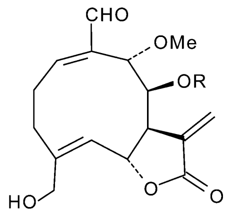
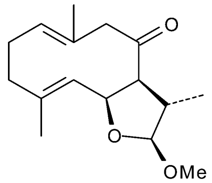


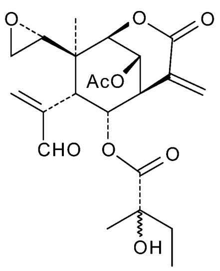
263 R = Ang
264 R = Meacr



265

15 Elemane

Zinagrandinolides A–C **266–268** are three new sesquiterpene lactones, which have been isolated from *Zinnia grandiflora*.[260] Two sesquiterpene glycosides sarcaglaboside C **269** and sarcaglaboside D **270** have been found in an extract from *Sarcandra glabra*.[246] A known furanoelemene sesquiterpene has been obtained from the marine octocoral *Muricea austera*.[233] The reaction of elemol with acetic acid–perchloric acid has been studied.[261]



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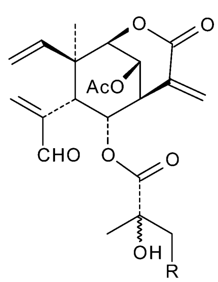
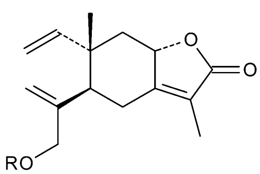


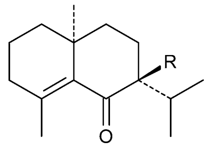
267 R = Me
268 R = H



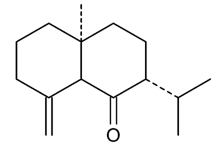
269 R = Glc
270 R = Glc-(6'-1'')-Api

16 Eudesmane, axane, cycloaxane, iphionane, isoiphionane and lindenane

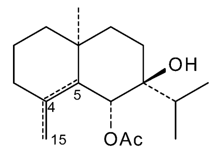
The eudesmane sesquiterpenes, isolated from the Asteraceae family, have been reviewed.[262] The essential oils from the liverwort *Plagiochila bifaria*[263] contain the new *ent*-eudesmenones **271–273**. Three eudesmane sesquiterpenes **274**, **276** and **278** have been obtained from another liverwort *Lepidozia fauriana*.[264] In this work, the structures of another two metabolites have been revised to **275** and **277**. These compounds had been obtained from *Bazzania tridens*[265] and *Lepidozia vitrea*,[266] respectively. The new eudesmane derivative **279** has been found in an extract from a marine *Streptomyces* species.[267]



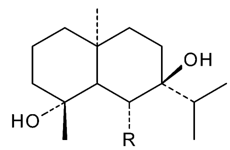
271 R = OH
272 R = H



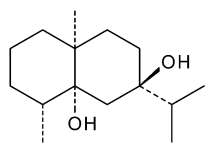
273



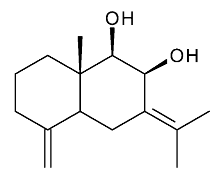
274 $\Delta^{4(5)}$
275 $\Delta^{4(15)}$



276 R = OAc
277 R = H

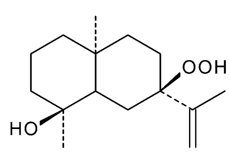


278



279

The new hydroperoxide **280** has been isolated from the aerial parts of *Aster spathulifolius*.[268] Two cytotoxic sesquiterpenes, **281** and **282**, with a eudesmane and an axane skeleton, respectively, have been found in *Jatropha neopauciflora*.[269] This plant also contains two cycloaxane derivatives, **283** and **284**.[140] A phytochemical study of a variety of *Juniperus polycarpus*[270] afforded two new eudesmane sesquiterpenes, **285** and **286**. Other compounds of this type, pterodontriol E **287** and pterodontriol F **288**, have been obtained from the aerial parts of *Laggera pterodonta*,[271] whilst plenoxide **289** has been isolated from *Schisandra plena*.[272] Another two new sesquiterpenes, **290** and **291**, have been obtained from



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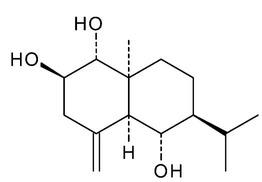

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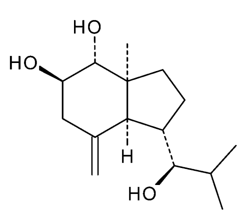
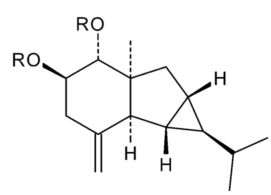
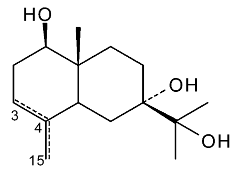


282

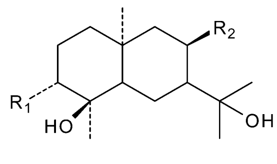


283 R = Ac
284 R = H



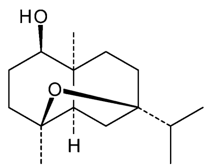
285 Δ^3

286 $\Delta^{4(15)}$

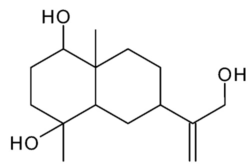


287 R_1 = OH R_2 = H

288 R_1 = H R_2 = OH



289



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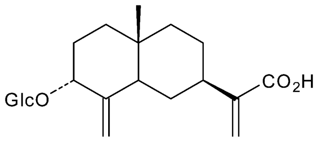
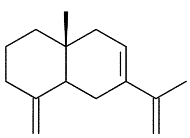


291

Sambucus williamsii,[273] and *Youngia japonica*,[26] respectively. The isolation of the new metabolites **292** and **293** from *Atractylodes macrocephala*[274] and **294** from *Chloranthus henryi*[275] has been reported. The structure of the sesquiterpene eudesma-5,12-dien-13-oic acid has been determined by X-ray analysis. This compound has been obtained from *Laggera pterodonta*.[276] The eudesmane glycosides **295** and **296** have been found in *Fissistigma pallens*[277] and *Pteris multifida*,[278] respectively, whilst an acetylated eudesmane glucoside **297** has been isolated from *Apodytes dimidiate*.[279] The antibacterial, antioxidant and α-amylase inhibitory activity of 3-oxo-selina-4,11(13)-dien-12-oic acid have been evaluated. This sesquiterpene has been obtained from *Varthemia iphionoides*.[280,281]



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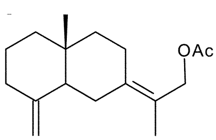

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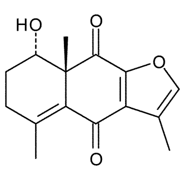

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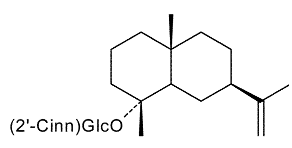

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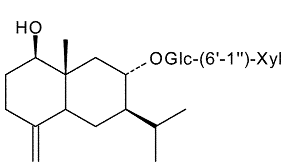

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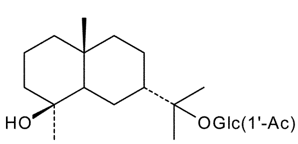
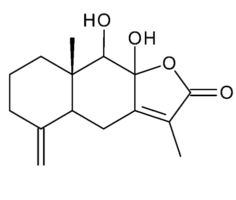


297

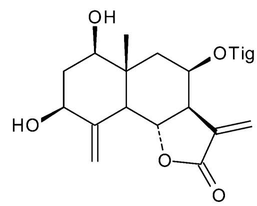
The molecular cloning, expression and mechanistic study on the germacradienol/geosmin synthase from *Streptomyces avermitilis* have been described.[282] Moreover, it has been shown that a germacradienol/germacrane D synthase from *Streptomyces coelicolor* converts farnesyl diphosphate into geosmin.[283] The first total syntheses of (±)-corymbolone[284] and of 6β-cinnamoyloxy-1α-hydroxy-5,10-bis-*epi*-eudesm-4-en-3-one[285] have been reported. The insect antifeedant activity of (+)-pterocarpol has been evaluated.[286]

Several dihydroagarofuran derivatives have been obtained from *Austroplenckia populnea*,[287] *Celastrus angulatus*,[288] *Celastrus orbiculatus*,[289] *Celastrus paniculatus*,[290] *Euonymus nanoides*,[291] *Microtropis fokienensis*,[292] *Pleurostylia opposita*,[293] *Reissantia buchananii*[294] and *Tripterygium wilfordii*.[295] Insights into the molecular mechanism of action of this type of sesquiterpenes, as specific non-transported inhibitors of human P-glycoprotein, have been described.[296]

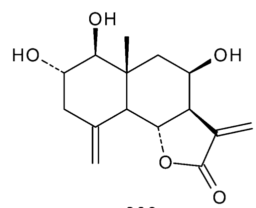
The new eudesmanolides which have been obtained from different species are listed in Table 2. Their structures have been shown to be **298–316**.



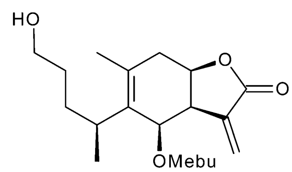
298 Atractylenolide V



299 Eupakirunsin H



300



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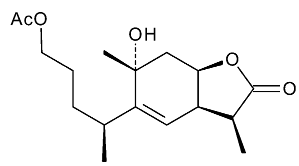
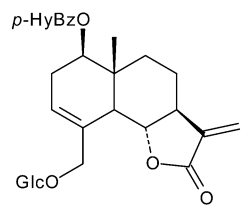


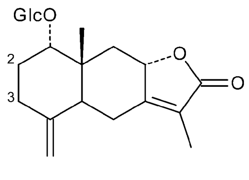
302



303

Table 2 Sources of eudesmanolides

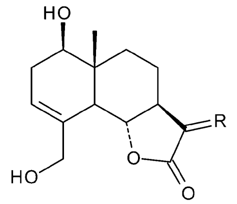
Source	Eudesmanolides	Ref.
Atractylodes macrocephala	**298**	274
Eupatorium kiirunense	**299**	241
Eupatorium lindleyanum	**300**	249
Inula britannica	**301**, **302**	244
Lantuca indica	**303**	297
Sarcandra glabra	**304**, **305**	246
Sonchus uliginosus	**306**, **307**, See text	298
Thapsia nitida	**308–316**	230



Sarcaglabosides A and B

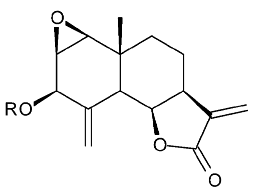
304

305 Δ^2



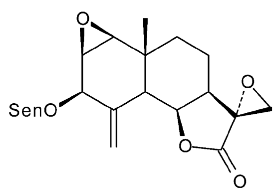
306 R = α-Me,H

307 R = CH_2



308 R = Sen

309 R = Val^i



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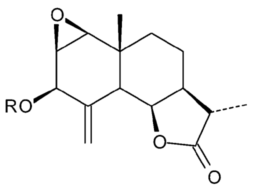
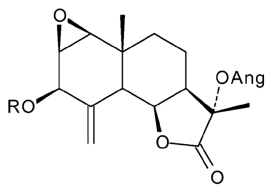


311 R = Sen

312 R = Ang

313 R = Val^i



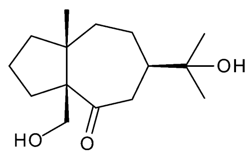
314 R = Sen

315 R = Ang

316 R = Val^i

Six new lactones have been obtained from *Sonchus uliginosus*.[298] Two are given in Table 2, but the other four had previously been isolated from *Sonchus transcarpicus*. Their structures were reported in our last review.[116] Biotransformations of the lactones α-santonin and 6β-santonin by the fungus *Abisidia coerulea* and by cell cultures of the plants *Asparagus officinalis* have been carried out.[299] The structure and biological activity of several chloro derivatives of α-santonin have been reported.[300] An enantioselective total synthesis of the antileukaemic secoeudesmane lactones (−)-eriolanin and (−)-eriolangin has been accomplished.[301] The acid catalysed isomerisation of several alantolactone derivatives has been studied.[302]

A rearranged eudesmane sesquiterpenoid **317**, with a novel bicyclic ring system, has been obtained from *Jasonia montana*.[303] This compound was named jasomontanone, but unfortunately a norsesquiterpene, also isolated from this species, had previously been given the same name.[304] This plant also contains the sesquiterpene 11,15-dihydroxy-isoiphionane-4-one **318**, which has been named montanone.[305] A previous version of this work had been published, and considered in our last review.[116] We think that the metabolite **317** is identical with montanone, because the physical and spectroscopic data reported for both compounds, **317** and **318**, are very similar. In consequence one of these structures must be erroneous, although the NMR data are more in accordance with the structure **317**. The isoiphionane sesquiterpene isofaurinone **319** has been found in the essential oil of *Anthemis aciphylla*.[306] A formal synthesis of the iphionane derivative **320** has been achieved.[307] The lindelanolide **321** has been isolated from *Sarcandra glabra*,[246] while a lindenane dimer, chloromultilide A **322**, has been obtained from *Chloranthus multistachys*.[308]



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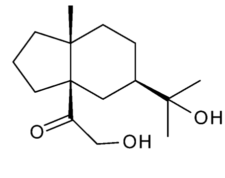

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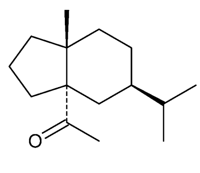

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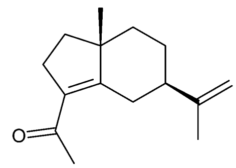

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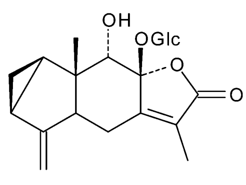

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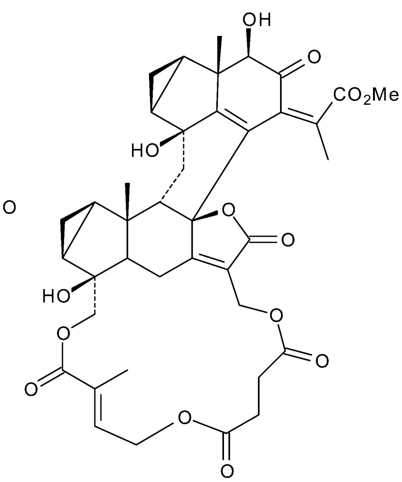
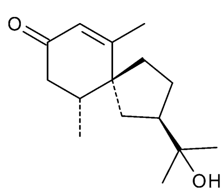


322

17 Vetisperane and spiroaxane

The new bioactive spirovetivane sesquiterpene **323** has been obtained from a Vietnamese agarwood.[309] A racemic total synthesis of α-vetisperane, hinesol and β-vetivone has been described,[310] whilst an enantiospecific total synthesis of (+)-solanascone, (+)-dehydrosolanascone and (+)-anhydro-β-rotunol has been accomplished.[311] (+)-2β-Hydroxysolanascone, the aglycone of the phytoalexin isolated from flue-cured tobacco leaves, has been synthesised.[312] The stereochemistry and the deuterium isotope effects associated with the cyclisation and rearrangements of (*E*,*E*)-farnesyl diphosphate, catalysed by hyoscyamus premnaspirodiene synthase (HPS), to give premnaspirodiene **324** have been described.[313]



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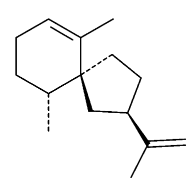
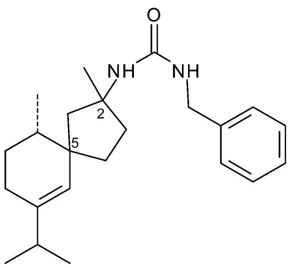
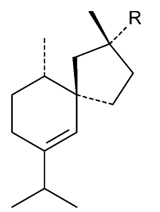


324

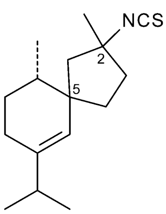
A new pair of diastereomeric spiroaxene derivatives, **325** and **326**, has been isolated from both sponges *Amorphinopsis foetida* and *Axinyssa aplysinoides*, collected at Papua New Guinea and Vanuatu, respectively. The first sponge also contains the known metabolite **327**, whose stereochemistry has now been revised, whilst the second contains the spiroaxene **328**, whose stereochemistry has also been corrected, and its new diastereomer **329**.[314] Another compound of this type, 3-oxo-axisonitrile-3 **330**, has been found in a Chinese sponge of the *Acanthella* genus.[315]



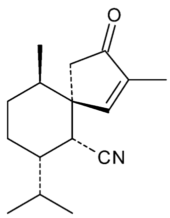
325 (2*S*,5*S*) or (2*R*,5*R*)
326 (2*S*,5*R*) or (2*R*,5*S*)



327 R = NHCHO
328 R = NCS



329 (2*S*,5*R*) or (2*R*,5*S*)

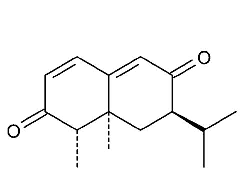


330

18 Eremophilane and bakkane

Pleodendione **331** and remophilanetriol **332** are two new eremophilane sesquiterpenes, which have been obtained from *Pleodendron costaricense*[45] and *Rehmannia glutinosa*,[316] respectively. The genus *Ligularia* is a good source of eremophilane derivatives. The sesquiterpenes **333** and **334** have been isolated from *Ligularia hodgsonii*[317] and *Ligularia myriocephala*,[318] respectively, whilst the compounds **335–336** and virgaurenones A–D **337–340** have been found in the varieties *oligocephala*[319] and *virgaurea*,[320] respectively, of *Ligularia virgaurea*. Chromatography of an extract from the roots and aerial parts of *Senecio mairetianus*[321] afforded the eremophilane derivative **341**. The Japanese butterbur (*Petasitis japonicus*) contains fukinoside A **342**, a novel sesquiterpene glycoside sulfate with anti-allergic activity.[322] Two chemotypes of *Petasites hybridus* are known; one produces petasin **343** whilst the other contains furanopetasin **344**, but only the first is useful for phytopharmaceutical preparations. Experimental crossings of these chemotypes have been performed to study the genetic basis of the occurrence of these sesquiterpenes.[323]

The furanoeremophilane **345** has been obtained from *Senecio kaschkarovii*.[324] A phytochemical study of *Roldana angulifolia*[325]



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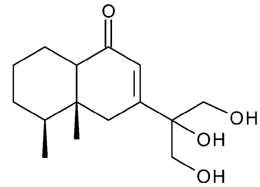

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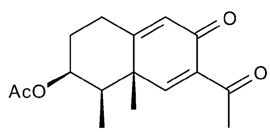

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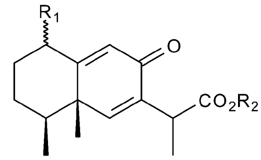
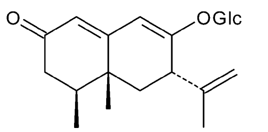


334 R_1 = β-OAng R_2 = Me
335 R_1 = α-OH R_2 = H



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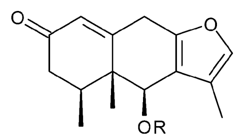
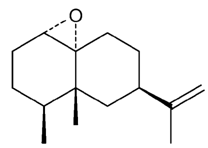


337 R = Ang
338 R = Ac
339 R = H
340 R = Ang(4'-OH)



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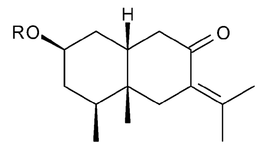
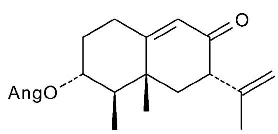


342 R = Glc(6'-SO_3K)



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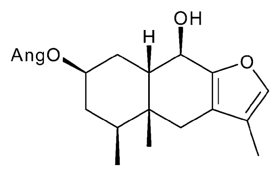
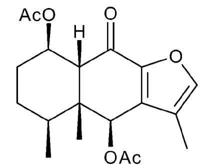


344

afforded the new sesquiterpenes angulifolide **346** and angulifolins A–C **347–349**. The structures of peribysin C and peribysin D have been revised to **350** and **352**, respectively.[326] These compounds had been isolated from a strain of *Periconia byssoides*, which was originally separated from the sea hare *Aplysia kurodai*.[327] Another strain of *Periconia* sp.,[328] isolated from the same sea hare, contains a further two new eremophilane derivatives, which have been named peribysin H **351** and peribysin I **353**.

The new eremophilanolides which have been isolated from different species are listed in Table 3. Their structures have been shown to be **354–381**.

The stereochemistry and the deuterium isotope effects associated with the cyclisation and rearrangements of (*E*,*E*)-farnesyl



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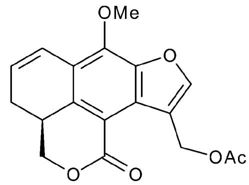

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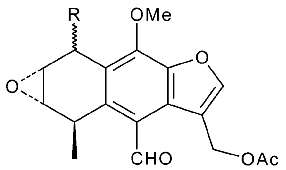
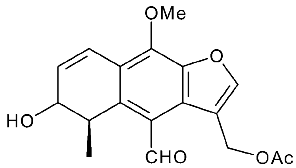


347 R = α-OH
348 R = β-OH



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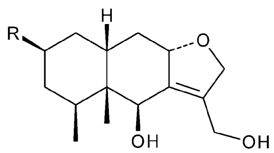
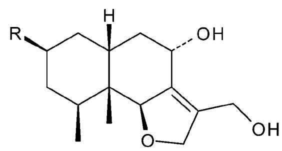


350 R = H
351 R = OH



352 R = H
353 R = OH

diphosphate, catalysed by tobacco epiaristolochene synthase (TEAS), to give the corresponding hydrocarbon have been investigated.[313] GC-MS methods have been used to make a detailed analysis of other sesquiterpenes formed in this cyclisation.[338] It has previously been shown that the side chain of the phenylalanine-178 of aristolochene synthase promotes the conversion of a eudesmane cation to aristolochene. Now, it has been reported that this catalytic function is mainly due to the large size of this chain, which facilitates the hydride shift from C-2 to C-3.[339]

Table 3 Sources of eremophilanolides

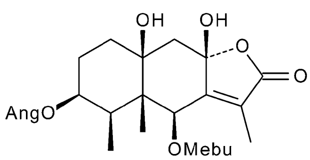
Source	Eremophilanolides	Ref.
Ligularia hodgsonii	**354–356**	317
Ligularia lapathifolia	**357–359**	329, 330
Ligularia muliensis	**360**, **362**	331, 332
Ligularia myriocephala	**363–366**	318, 333
Ligularia platyglossa	**367**	334
Ligularia virgaurea var. *oligocephala*	**368**, **369**	335
Ligularia virgaurea var. *virgaurea*	**370**, **371**	320
Senecio burtonii	**372**	336
Senecio mairetianus	**373–380**	321
Senecio poepigii	**381**	337

The photosensitised oxidation of several furanoeremophilane sesquiterpenes has been investigated by two different groups.[340,341] (−)-Carvone has been used as starting material in a synthesis of (−)-aristolochene,[342] whilst a racemic synthesis of 6-hydroxyeuryopsin, 1,10-epoxy-6-hydroxyeuryopsin, toluccanolide A and toluccanolide C has been reported.[343] The structures of several compounds produced in the basic hydrolysis of four eremophilane esters, which were isolated from *Robinsonecio gerberifolius*, have been assigned.[344]

A synthesis of (±)-bakkenolide A has been described,[345] whilst an approach to the diastereoselective construction of the *nor*-bakkane skeleton has been reported.[346]

19 Guaiane, pseudoguaiane, xanthane and patchoulane

The new azulene pigments **382** and **383–384** have been obtained from the fruiting bodies of *Lactarius deliciosus*[347] and *Lactarius hatsudake*,[348] respectively. Another compound of this type **385**, a known bioactive chamazulene derivative, has been found to be formed from the lactone matricin by treatment with artificial gastric fluids.[349] The essential oil of *Ajania fruticulosa* contains the bisazulene **386**, whose structure was determined by X-ray analysis.[350]



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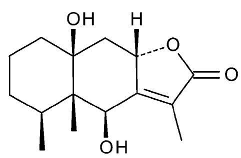

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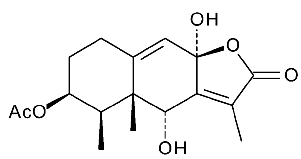

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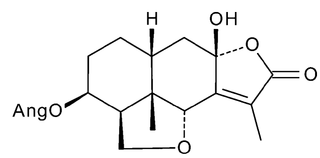

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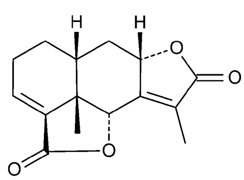

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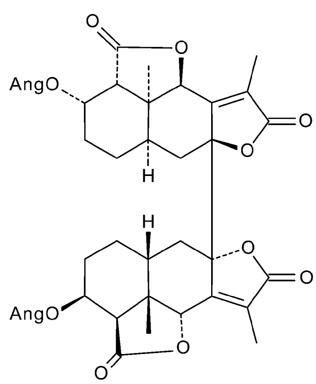

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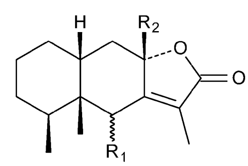
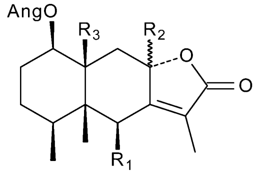
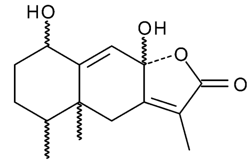


360 R_1 = β-OAc R_2 = OH
361 R_1 = α-OH R_2 = H
362 R_1 = α-OAc R_2 = H



363 R_1 = OH R_2 = β-OMe R_3 = β-OH
364 R_1 = OH R_2 = α-OMe R_3 = α-OH
365 R_1 = OAng R_2 = β-OH R_3 = β-OH
366 R_1 = OAng R_2 = α-OH R_3 = α-OH



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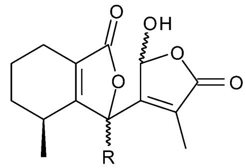
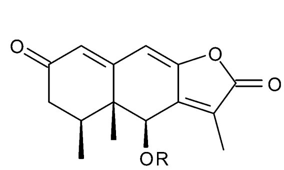
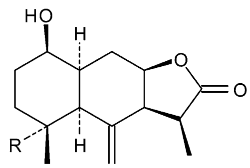


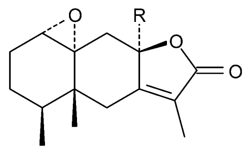
Ligulides C_1 and C_2
368 R = α-Me
369 R = β-Me



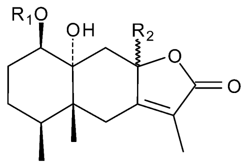
Virgaurenolides A and B
370 R = Ang
371 R = Ac



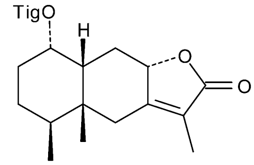
372 R = Meacr(4'-OH)



373 R = H
374 R = OH



375 R_1 = Me R_2 = α-OMe
376 R_1 = Me R_2 = α-OH
377 R_1 = Me R_2 = α-H
378 R_1 = H R_2 = α-H
379 R_1 = H R_2 = α-OH
380 R_1 = H R_2 = β-OH



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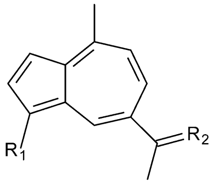
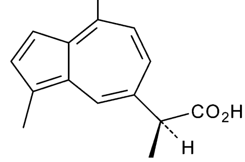


382 R_1 = CO_2H R_2 = O
383 R_1 = CHO R_2 = Me,OH
384 R_1 = CO_2H R_2 = Me,H



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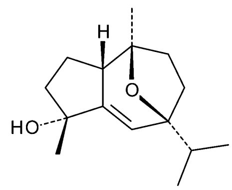

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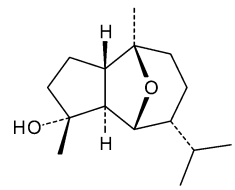

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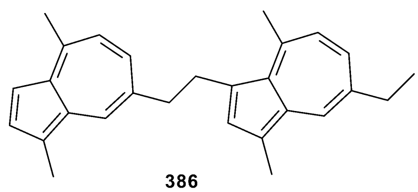

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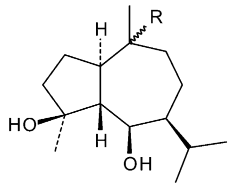
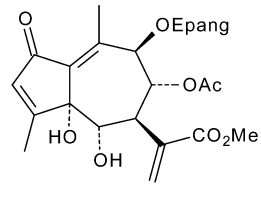


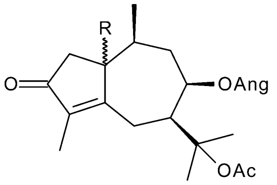
389 R = α-OH
390 R = β-OH



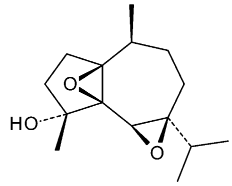
391

Pubinernoid B **387** is a new guaiane derivative, which has been obtained from *Schisandra pubescens*.[143] A phytochemical study of the aerial parts of *Crysothamnus viscidiflorus* afforded a sesquiterpene **388**, which was named chrysothol.[351] The authors have indicated that the spectroscopic data of this compound were identical with those reported for buchariol, isolated from *Salvia bucharica*;[352] consequently the structure of the latter must be corrected. In addition, it is also shown in this work that the structure given to another sesquiterpene, obtained from *Fagonia boveana*,[353] is erroneous. The structures **389** or **390** were proposed. The exudates of the aerial parts of *Balsamorhiza sagittata* and *Balsamorhiza macrophilla*[354] contain the new guaiane derivative **391**. A study of a methanolic extract from the fruits of *Torilis japonica*[355] afforded the cytotoxic sesquiterpenes torilin **392**, 1α-hydroxy-torilin **393** and the new metabolite 1β-hydroxy-torilin **394**. It has also been shown that torilin may be an ideal anti-arrhythmic drug for atrial fibrillation.[356] The bioactive guaiane derivatives **395** and **396** have been identified as constituents of a soft coral, *Sinularia* sp.[357]

A total synthesis of the cytotoxic guaiapyridine sesquiterpene alkaloid (+)-cananodine **397** has been accomplished.[358] The norsesquiterpene (−)-clavukerin A has been synthesised using a bioinspired procedure.[359] The first total synthesis of (+)-alismoxide and (+)-4-*epi*-alismoxide has been achieved.[360] In this work the structure of a natural guaienediol, isolated from *Silphium*



392 R = β-H
393 R = α-OH
394 R = β-OH



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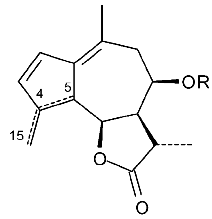
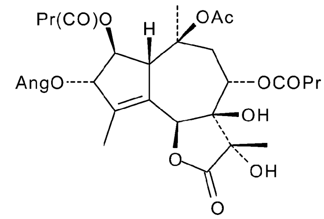
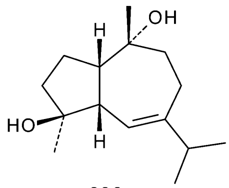


442 $\Delta^{4(5)}$ R = Ac
443 $\Delta^{4(5)}$ R = Ang
444 $\Delta^{4(15)}$ R = Ac



Thapsivillosin L **445**



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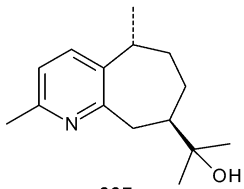

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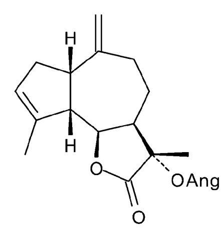

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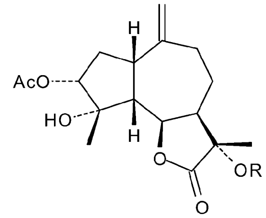
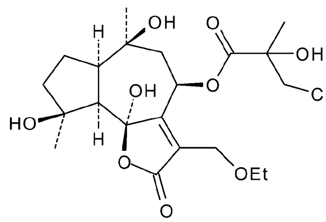
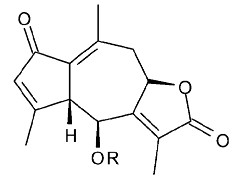


447 R = Tig
448 R = Ang



Vernocinolide A **449**



Nubenolide **450** R = H
451 R = Ac

perfoliatum, has also been corrected to that of the known sesquiterpene (−)-alismoxide. The microbiological transformation of (−)-guaiol by the fungus *Eurotium rubrum* has been investigated.[361] Synthetic approaches to hydroazulenes and guaianes have been reported.[362]

Many new guaianolides have been isolated during the period covered by this review (see Table 4). The novel guaian-6a,12-olides, *i.e.* **398–441**, are listed in Table 5 and other new guaianolides are represented by the 6β,12-lactones **442–449**, the guaian-8,12-olides **450–451** and the dimers **452–457**.

The lactone hololeucin **429**, isolated from *Centaurea hololeuca*, represents the first example of a natural cyclic carbonate.[364] Known guaianolides have been obtained from *Centaurea helenioides*,[375] *Centaurea ptosimopappa*,[376] *Centaurea scabiosa*,[377] *Chondrilla juncea*,[378] *Ixeris dentata*,[379] *Rhaponticum pulchrum*[380] and *Taraxacum bessarabicum*[381] The structures of a guaianolide[382] **458** and of a tetrabromoderivative of cyclopropyldihydroargablin[383] have been determined by X-ray analysis. A conformational analysis of several 11,13-oxetane lactones has been carried out.[384] The absolute configuration of the guaianolide **459** has been unambiguously determined by comparison of the calculated and experimental vibrational circular dichroism (VCD) spectra.[385]

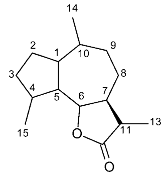
Table 4 Sources of guaianolides

Source	Guaianolides	Ref.
Achillea asplenifolia	**398**, **405–408**, **442**, **443**	237
Achillea clavennae	**436–439**	79
Achillea collina	**399–402**, **404**, **409**, **414**, **444**, **452**	238
Amphoricarpos neumayeri	**403**, **431–434**, **440**	363
Balsamorhiza sagittata	**411**, **412**	354
Centaurea hololeuca	**429**	364
Cichorium endivia	**416**	365
Crepis cameroonica	**422**	366
Eupatorium chinense	**417–419**, **425**, **426**, **441**	240
Helianthus annuus	**415**	243
Inula britannica	**453–456**	367
Ixeris chinensis	**424**, **435**	368
Ixeris sonchifolia	**410**	369
Laurus nobilis	**423**	370
Mulgedium tataricum	**413**	245
Salvia nubicola	**450**, **451**, **457**	371
Tanacetum fruticulosum	**427**, **428**	372
Thapsia garganica	**445**	373
Thapsia nitida	**446–448**	230
Vernonia cinerea	**449**	374
Youngia japonica	**420**, **421**, **430**	26

The influence of the cultivation site on the guaianolide composition of *Cichorium intybus* has been studied.[386] Some of the lactones isolated from *Eupatorium capillifolium* showed potent inhibitory effects on the growth of HeLa cells,[387] whilst cynaropicrin has been shown to be a potent and irreversible inhibitor of the antibacterial target enzyme MurA.[257] The biotransformation of dehydrocostuslactone by the fungus *Mucor polymorphosporus* has been investigated.[388] An enantioselective synthesis of a 7,11-dihydroxyguaianolide, possessing the stereochemistry of thapsigargin, has been described.[389] A study of a thapsigargin derivative has revealed the importance of the length and flexibility of the side chain attached to O-8 in the activity of this sesquiterpene lactone, and its analogues, towards prostate cancer cells.[390]

Five novel pseudoguaianolides, dichrocepholides A–E **460–464**, have been isolated from *Dichrocephala integrifolia*.[391] Another two new compounds of this type, deacetyltetraneurin A **465** and hysterone E **466**, have been found in an extract from the flowers of *Parthenium hysterophorus*.[392] An enantioselective synthesis of (+)-8-*epi*-xanthatin has been reported.[393]

Table 5 Novel guaian-6α,12-olides



Name	Structure	Position of double bond(s)	Substituents and configurations	Ref.
Solidaginolide A isomer	**398**		1α,2α-epoxy, 4α,5α-epoxy, 8α-OAng, 10β-OH, 11α	237
Solidaginolide A isomer	**399**		1α,2α-epoxy, 4α,5α-epoxy, 8α-OTig, 10β-OH, 11α	238
10-*epi*-Ezoartemin derivative	**400**		1β,2β-epoxy, 3β,4β-epoxy, 8α-OTig, 10β-OH, 11α	238
10-*epi*-Solidaginolide A	**401**		1α,2α-epoxy, 3α,4α-epoxy, 8α-OTig, 10β-OH, 11α	238
10-*epi*-Solidaginolide A deriv.	**402**		1α,2α-epoxy, 3α,4α-epoxy, 8α-OAng, 10β-OH, 11α	238
Amphoricarpolide deriv.	**403**		4α, 10α,14-epoxy, 11α-OH, 13-OH	363
Artabsinolide A deriv.	**404**	1–5	2-oxo, 4α-OH, 8α-OTig, 10β-OH, 11α	238
Tannunolide B derivative	**405**	2–3, 4–5, 10–1	8α-OAc (or -OAng), 11α	237
Tannunolide C derivative	**406**	2–3, 4–15, 10–1	8α-OAng, 11α	237
Matricin derivative	**407**	2–3, 10–1	4α-OMe, 8α-OAng, 11α	237
Matricin derivative	**408**	2–3, 10–1	4β-OH (or -OMe), 8α-OAng, 11α	237
Tanaparthin derivative	**409**	2–3	2α,4α-dioxo, 8α-OCOPri (or -OTig), 10β-OH, 11α	238
Ixerin Z$_A$	**410**	3–4, 10–1, 11–13	2-oxo, 3-OGlc(6′-*p*-AcOBz)	369
Zubergenin derivative	**411**	3–4, 10–1, 11–13	8α-OAc, 9β-OEpang	354
Zubergenin derivative	**412**	3–4, 10–1, 11–13	5α-OH, 8α-OAc, 9β-OAng	354
Lactucin derivative	**413**	3–4, 10–1, 11–13	2-oxo, 8α-OR$_1$, 15-OH	245
5α-Hydroxymatricarin	**414**	3–4, 10–1	2-oxo, 5α-OH, 8α-OAc, 11α	238
Annuolide H	**415**	3–4, 11–13	2α-OH, 8β-OAng, 10α-OH	243
Hieracin derivative	**416**	3–4	2-oxo, 8α-OH, 10β-OMe, 11α, 15-OH	365
Eupatochinilide V	**417**	4–5, 10,1, 11–13	3β-OH, 8β-OAng, 14-oxo	240
Eupatochinilide VI	**418**	4–5, 10,1, 11–13	3β-OH, 8α-OAng, 14-oxo	240
Eupatochinilide VII	**419**	4–5, 10,1, 11–13	3β-OH, 8β-OEpang, 14-oxo	240
Zulazanin C derivative	**420**	4–15, 10–14, 11–13	3β-OGlc(3′-OR$_1$)	26
Zulazanin C derivative	**421**	4–15, 10–14, 11–13	3β-OGlc(3′-OR$_1$), 8α-OH	26
8-Desacylcynaropicrin	**422**	4–15, 10–14, 11–13	3β-OH, 9β-OH	366
Zaluzanin C derivative	**423**	4–15, 10–14, 11–13	3β-Cl	370
Chinensiolide E	**424**	4–15, 11–13	3-OR$_2$, 10α-OH	368
Eupatochinilide II	**425**	10–1, 11–13	3α,4α-epoxy, 8β-OEpang, 14-OH	240
Eupatochinilide IV	**426**	10–1, 11–13	3β-Cl, 4α-OH, 8β-OAng, 14-OH	240
Carlaolide A	**427**	10–1, 11–13	3β,4β-epoxy, 8α-OCOPri	372
Carlaolide B	**428**	10–1, 11–13	3β,4β-epoxy, 8α-OAng	372
Hololeucin	**429**	10–14, 11–13	3β,4β-carbonate, 8α-OMeacr(4′-OH), 15-OH	364
Zaluzanin C derivative	**430**	10–14, 11–13	3-oxo, 4α, 8α-OR$_1$	26
Amphoricarpolide deriv.	**431**	10–14, 11–13	3β-OH, 4α, 9β-OH, 15-OAc	363
Amphoricarpolide deriv.	**432**	10–14, 11–13	4α, 15-OAc	363
Amphoricarpolide deriv.	**433**	10–14	4α, 11α-OH, 13-OH, 15-OH (or -OAc)	363
Amphoricarpolide deriv.	**434**	10–14	4α, 11α-OH, 13-Cl, 15-OAc	363
Chinensiolide D	**435**	11–13	3-oxo, 4β, 10α-OH	368
9α-Acetoxyartecanin	**436**	11–13	1β,2β-epoxy, 3β,4β-epoxy, 9α-OAc, 10α-OH	79
A chlorohydrin	**437**	11–13	1β,2β-epoxy, 3α-Cl, 4β-OH, 10α-OH	79
A chlorohydrin	**438**	11–13	1β,2β-epoxy, 3α-Cl, 4β-OH, 9α-OAc, 10α-OH	79
9α-Acetoxyandalucin	**439**	11–13	1α,2α-epoxy, 3α-OH, 4β-Cl, 9α-OAc, 10α-OH	79
Amphoricarpolide deriv.	**440**	11–13	4α, 10α,14-epoxy, 15-OH	363
Eupatochinilide III	**441**	11–13	3α,4α-epoxy, 8β-OTig(4′-OH), 10α,14-epoxy	240

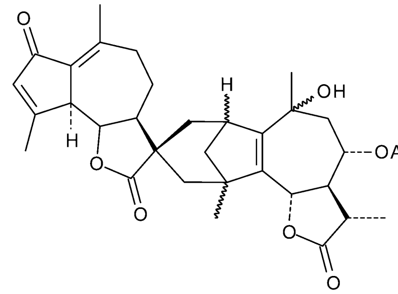
R$_1$ = *p* -CO–CH$_2$-(C$_6$H$_4$)OH R$_2$ = Glc(2′-OR$_1$, 6′-OR$_1$).

The diverse sesquiterpene profile of *Pogostemon cablin* has been correlated with the existence of a limited number of sesquiterpene synthases in this species.[394] Cyperotundone and α-cyperone have been obtained from dried tubers of *Cyperus rotundus*. The first of these ketones was oxidised with selenium dioxide to give 4-patchoulene-2,3-dione, which showed plant growth regulatory properties.[395] A new synthetic route to the patchoulol skeleton has been devised. The compounds obtained have permitted novel insights into the structural requirements of patchouli odorants.[396]

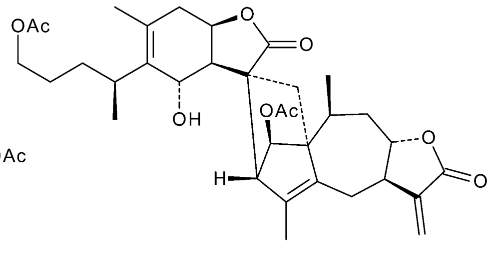
20 Aromadendrane, bicyclogermacrane, aristolane, sinulariolane and valerenane

The Formosan soft coral *Clavularia inflata* var. *luzoniana*[397] contains three new aromadendrane sesquiterpenoids **467–469**. The known sesquiterpene ledol has been obtained from the leaf surface of *Solanum tuberosum*.[232] A synthesis of (±)-epiglobulol has been reported.[398]

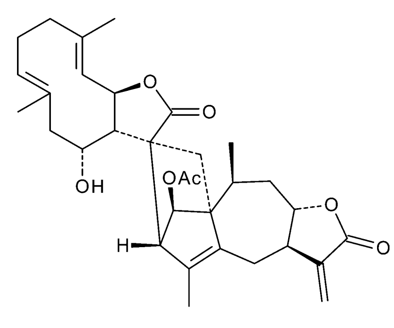
Kissoones A–C **470–472** and the compound **473** are four rearranged bicyclogermacrane derivatives, which have been isolated



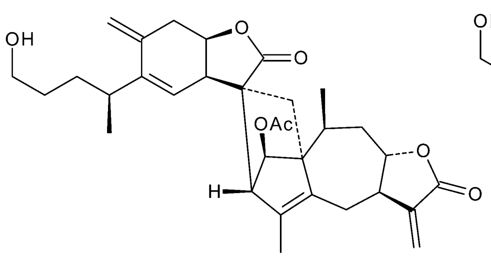
Achicollinolide **452**



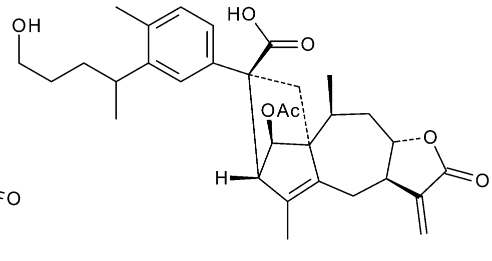
Inulanolide A **453**



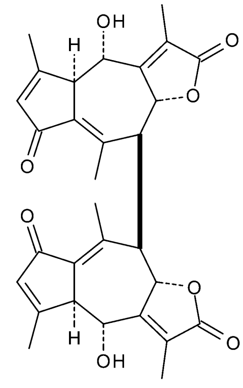
Inulanolide B **454**



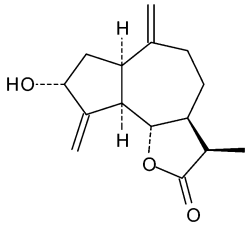
Inulanolide C **455**



Inulanolide D **456**



Bisnubenolide **457**



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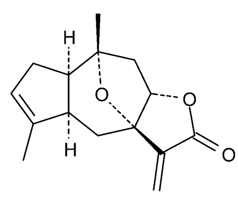

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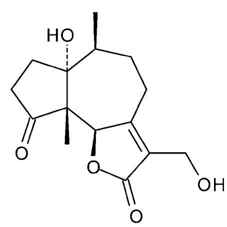

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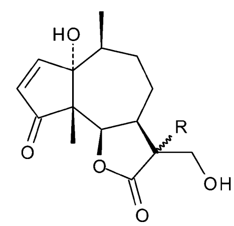
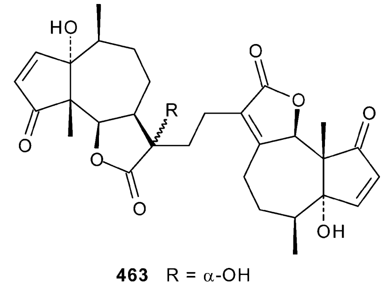


461 R = α-OH
462 R = β-OH

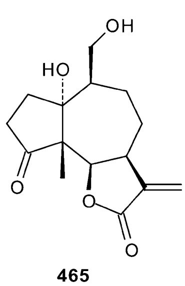
from the roots of *Valeriana fauriei*.[399,400] The biotransformations of the sesquiterpenes (+)-aristol-1(10)-ene and plagiochilide by three microorganisms, *Chlorella fusca* var. *vacuolata*, a *Mucor* species and *Aspergillus niger*, have been investigated.[401] Two novel valerenane sesquiterpenes, caulerpal A **474** and caulerpal B **475**, have been found in the Chinese green alga *Caulerpa taxifolia*.[402] The structure of another valerenane derivative sinularianin B has been determined as **476**. This sesquiterpenoid has been isolated from a Formosan soft coral of the *Sinularia* genus.[403] Other known sesquiterpenes of this type valerenal, valerenic acid and acetylvalerenolic acid have been obtained by a bioassay-guided fractionation of an extract from *Valeriana officinalis*. The two acids showed inhibitory activity against NF-κB in HeLa cells.[404]



463 R = α-OH
464 R = β-OH

21 Cyclomyltaylane

Five novel cyclomyltaylanoids **477–481** have been found in an extract from the Malagasy liverwort *Bazzania madagassa*.[405] Another liverwort, *Reboulia hemisphaerica*, collected in Japan, contains three new cyclomyltaylane derivatives **482–484** and the previously known cyclomyltaylan-5α-ol **485**. The absolute configuration of the latter has now been determined by application of the modified



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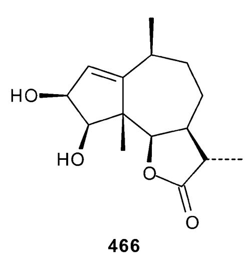

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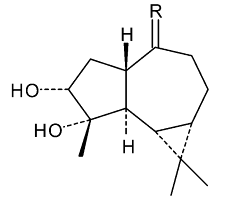
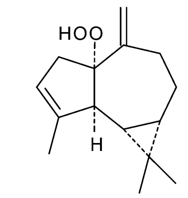


467 R = β-OH,Me
468 R = CH_2



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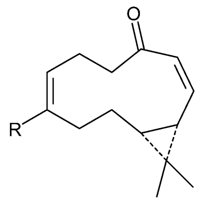
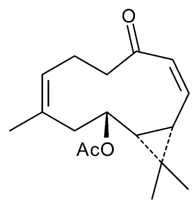


470 R = Me
471 R = CH_2OH
472 R = CH_2OAc



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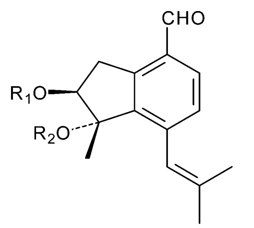
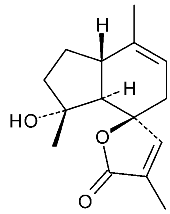
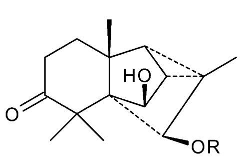


474 R_1 = Ac R_2 = H
475 R_1 = H R_2 = Me

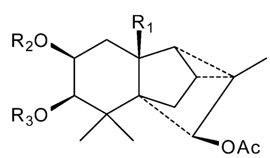


476

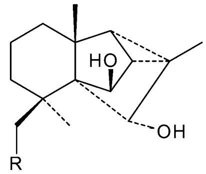
Mosher's method to one of the metabolites **486**, which was also obtained in this work by biotransformation of **485** with the fungus *Aspergillus niger*.[124]



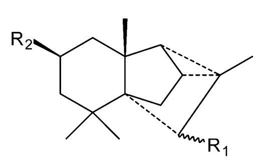
477 R = Ac
478 R = H



479 R_1 = Me R_2 = H R_3 = Ac
480 R_1 = CH_2OAc R_2 = H R_3 = Ac
481 R_1 = CH_2OAc R_2 = Ac R_3 = H



482 R = H
483 R = OAc



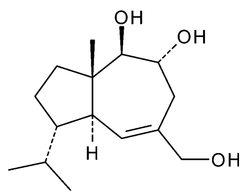
484 R_1 = β-OH R_2 = H
485 R_1 = α-OH R_2 = H
486 R_1 = α-OH R_2 = OH

22 Salvionane

A new "isodaucane" (salvionane) derivative **487** has been isolated from the bark of *Jatropha neopauciflora*.[140]

23 Miscellaneous sesquiterpenoids

The full paper with the structure of stereumone A has appeared.[406] Vibralactone **488** has been obtained from cultures of the basidiomycete *Boreostereum vibrans*. This C_{13} derivative inhibits pancreatic lipase.[407] The total synthesis of the fungal metabolites (+)-massarinolin B and (+)-4-*epi*-massarinolin B has been achieved.[408]



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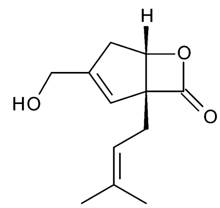
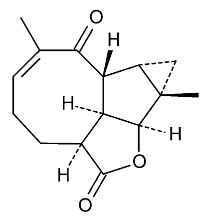


488

Naupliolide **489**, a sesquiterpene with a new carbon framework, has been isolated from *Nauplius graveolens*. The authors suggest that this metabolite may be formed from asteriscunolide C, also isolated from this plant.[409] The novel lactone **490** has been found in the study of an extract from *Laurus nobilis*.[370] An enantioselective synthesis of the (1*S*,5*R*)-enantiomer of litseaverticillol A and litseaverticillol B has been accomplished.[410] On the other hand, successful biomimetic syntheses of the litseaverticillol family of sesquiterpenes have been achieved, using singlet oxygen chemistry.[411] In this work the structure of litseaverticillol E has been revised to **491**. A total synthesis of racemic salsolene oxide **492** has been reported.[412]



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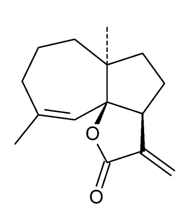

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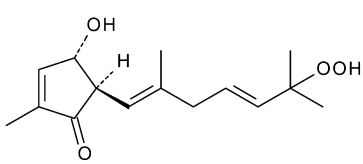

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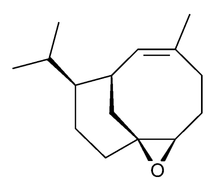
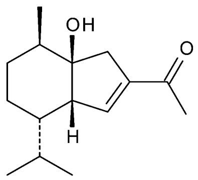
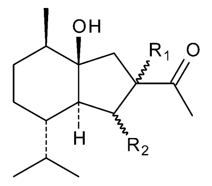


492

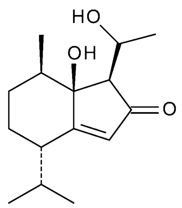
Five minor sesquiterpenes **493–497**, have been isolated from the brown alga *Dictyopteris divaricata*. These compounds have two new different carbon frameworks, which may be derived from a cadinane.[184] The Formosan soft coral *Clavularia inflata* var. *luzoniana*[397] contains the sesquiterpenoid **498**, which possesses a new ring system. Three sesquiterpenes with a novel carbon skeleton, paralemnanone **499**, isoparalemnanone, **500** and paralemnanol **501**, have been isolated from another soft coral, *Paralemnalia thyrsoides*.[413] The sesquiterpene sinularianin A **502**, which also has a new carbon framework named sinulariolane, has been isolated from a coral of the *Sinularia* genus.[403] The Okinawan marine sponge *Dysidea chlorea* contains four new sesquiterpenes, haterumadysins A–D **503–506**. These compounds inhibit the division of fertilised sea urchin eggs.[414] Spirofragilin **507** has been found in another species of this genus, *Dysidea fragilis*, which was collected in the South China Sea,[415] whilst *O*-methyl nakafuran-8 lactone **508** has been obtained from a Hainan sponge of the same genus.[416] Enantiospecific syntheses of 2-pupukeanone[417] and several allopupukeanones[418] have been achieved. Two new cytotoxic sesquiterpenoids, metachromin J **509** and metachromin K **510**, have been isolated from a marine sponge of the *Spongia* genus.[419]



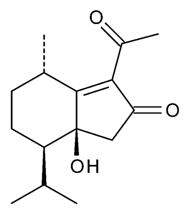
493



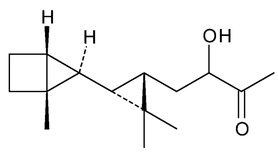
494 R_1 = α-H R_2 = β-OH
495 R_1 = β-H R_2 = α-OH



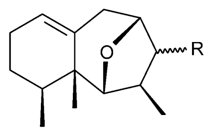
496



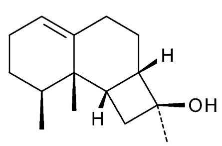
497



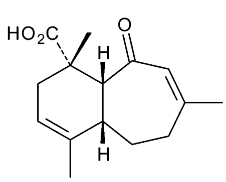
498



499 R = β-OH
500 R = α-OH



501



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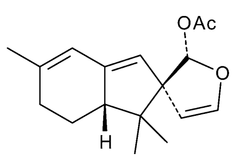
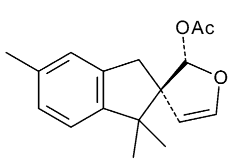
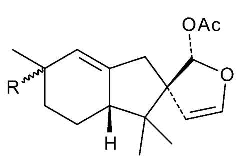


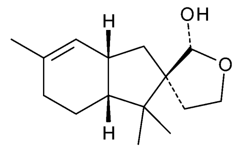
503



504



505 R = β-OOH
506 R = α-OOH



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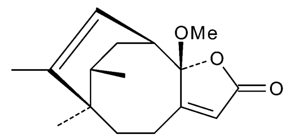
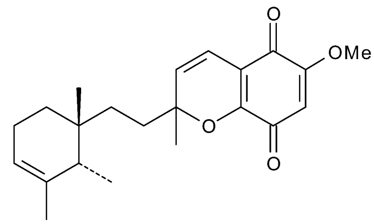
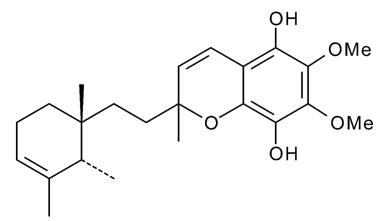


508



509



510

The germination of the seeds of some parasitic plants depends on the secretion of strigolactones. The occurrence, isolation, biosynthesis and the mode of action of these carotenoid-derived metabolites have been reviewed.[420,421]

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