

Eudesmane sesquiterpenoids from the Asteraceae family†

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This review covers the structures and biological activities of eudesmane-type sesquiterpenoids from the plants of the Asteraceae family. Biosynthetic studies or chemical syntheses leading to the revision of structures or stereochemistries have also been included, and 593 references are cited.

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1 Introduction

The large family Asteraceae (= Compositae) contains 25 000–30 000 species belonging to over 1000 genera. Many species have been used as sources of rubber, medicines, edible oils, vegetables,

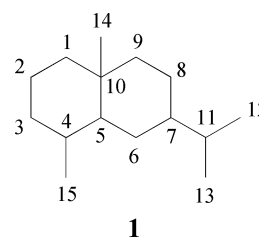
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pesticides and so on. Some are popular ornamental plants. The genera *Aster*, *Inula*, *Xanthium*, *Eupatorium*, *Carpesium*, *Saussurea* and *Taraxacum* are sources of various medicines; the whole plants of *Taraxacum kok-saghyz* and *Parthenium hysterophorus* are a source of rubber, and the seeds of *Helianthus annuus*, *Guizotia abyssinica* and *Xanthium sibiricum* are sources of edible oils. The extract from the leaves of *Blumea balsamifera* are used for the manufacture of borneol, and *Oxalis corymbosa* is used as a source of pyrethrum pesticides.¹ This family is a rich source of sesquiterpenoid natural products, especially those with the eudesmane framework.

The eudesmanoids are biosynthesised from farnesyl pyrophosphate,² and approximately 1000 natural eudesmanoids have been identified from the Asteraceae family, with many different oxygenation and cleavage patterns. The configuration and numbering of eudesmane (**1**, 2-isopropyl-4a,8-dimethyldecahydro-naphthalene) is shown below.



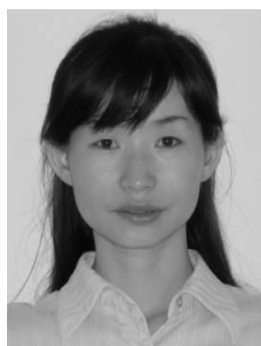
During the last two decades, eudesmane-type sesquiterpenoids and their biological activities from Asteraceae species have been the focus of numerous phytochemical, pharmacological and synthetic studies. Because the sesquiterpenoids exhibit a wide range of biological activities, and include compounds that are plant growth regulators, insect antifeedants, antifungals, anti-tumour compounds and antibacterials, there has been much interest in relating structure and oxygenation patterns to function.

This article reviews the structures and biological activities of novel eudesmane-type sesquiterpenoids from Asteraceae species. Studies involving biosynthesis and chemical synthesis and leading to a deeper chemical and biological understanding of these metabolites are discussed. The review highlights 972 compounds, and includes those with re-assigned structures or newly established stereochemistry where appropriate.

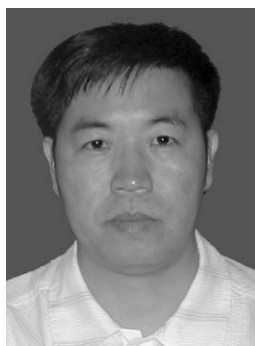
Quan-Xiang Wu received her M.Sc. and Ph.D. degrees in Organic Chemistry from Lanzhou University, China. Her research interests focus on natural products chemistry, including isolation and structural elucidation of biologically active secondary metabolites, especially in the field of terpenes from the higher plants. She obtained a permanent position in Lanzhou University as Senior Lecturer in 2005.

Yan-Ping Shi was born in Gansu (P. R. China) in 1965 and received his M.Sc. and Ph.D. degrees in Organic Chemistry from Lanzhou University, China in 1992 and 1996, respectively. He completed periods as a post-doctoral research associate in cell biology at Lanzhou University in 1998, and in marine natural products chemistry at the University of Puerto Rico in 2001, as well as being honoured with an Award from the Special Foundation for 'Bairen Jihua' at the Chinese Academy of Sciences (CAS) in 2000. He obtained a permanent position in the Lanzhou Institute of Chemical Physics (CAS) as a Research Professor in 2001, and was appointed Research Professor at Lanzhou University in 2003. Since 2004, he has been the Director of the Key Laboratory for Natural Medicine for Gansu Province. His research interests cover the chemistry of natural products, especially the isolation, purification, and structural elucidation of pharmacologically active terpenoids, together with determination of natural products in plant materials. He has authored over 100 scientific publications and was honoured with two 2nd Prizes for the Advancement of Science & Technology by the National Education Department of China in 1999 and 2005, respectively.

Zhong-Jian Jia was a Professor of Organic Chemistry at Lanzhou University, China, from 1959 to 2006. Her research interests focus on the chemistry of natural products from medicinal and toxic plants, mainly those of China. She serves on the Editorial Board of Asia Natural Products Research and is a member of the Evaluation Committee of Chinese Risk Investment of New Drug Development. She has authored more than 350 scientific publications and has been honoured with four 2nd Prizes and one 3rd Prize for the Advancement of Science & Technology by the National Education Department of China, and with one 3rd Prize for the Advancement of Science & Technology by the Sciences Commission of Gansu Province. She has also received the 'Gold Cattle' Prize from the National Development Program Commission (the Ministry of Education of China and the Ministry of Sciences and Techniques of China).



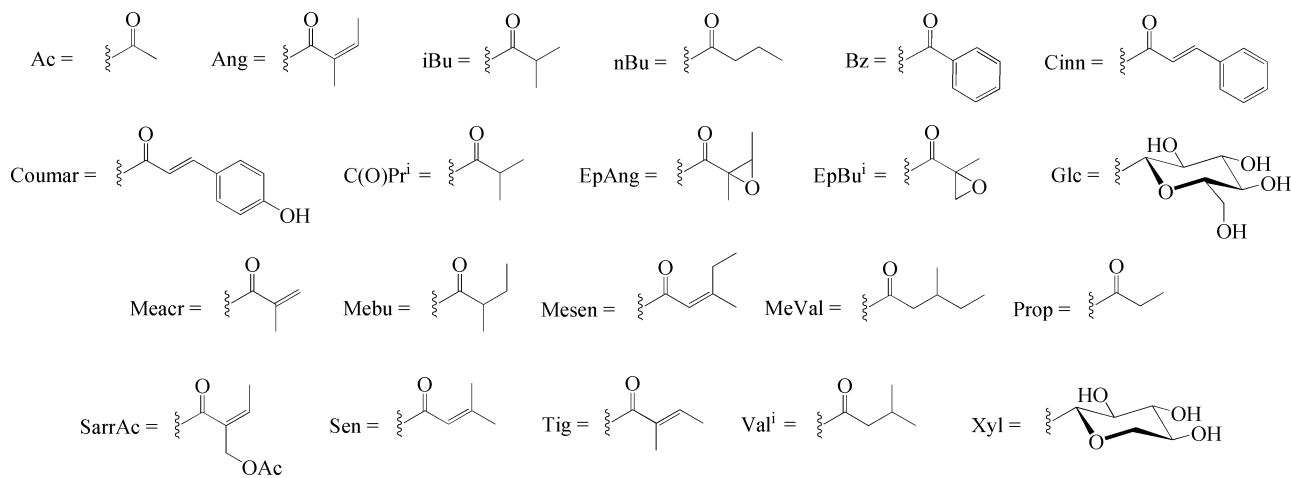
Quan-Xiang Wu



Yan-Ping Shi

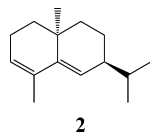


Zhong-Jian Jia



2 Eudesmenes

Non-oxygenated eudesmane sesquiterpenoids are hardly represented in the Asteraceae family. Eudesmene **2** was isolated from *Ursinia trifida*,³ and its isomer, 10-*epi*-eudesma-3,5-diene, has been prepared from santonin.⁴ The total syntheses of 4(15),7-dieneudesmane and (\pm)-vetiselinene have been reported.⁵



3 Oxygenated eudesmanes

Oxygenated eudesmanes form the major class of sesquiterpenoids in Asteraceae species, and include alcohols, ethers, epoxides, peroxides, aldehydes, ketones, carboxylic acids and lactones. The different functional groups are important in determining the individual biological activities of the various sesquiterpenoids. These compounds are discussed in more detail below.

3.1 Hydroxyl-substituted eudesmanes

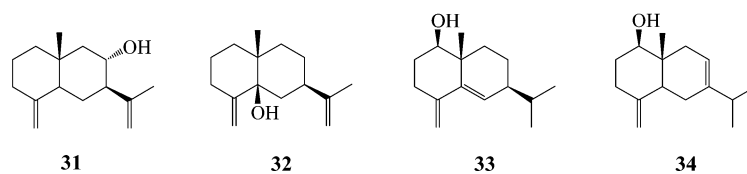
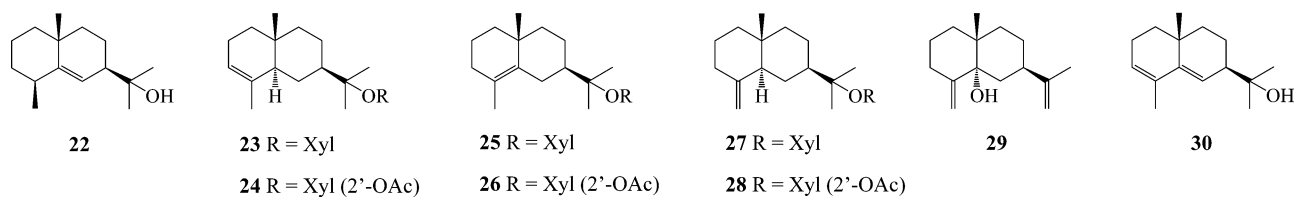
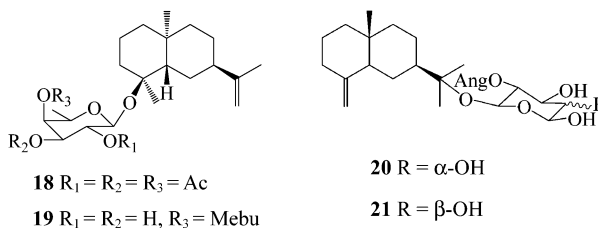
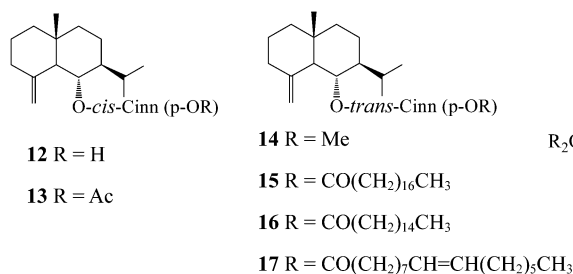
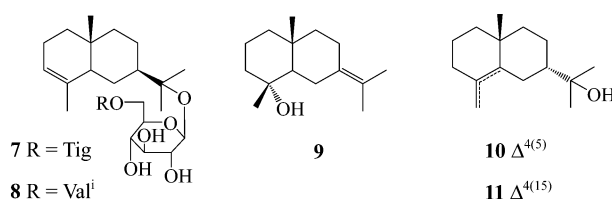
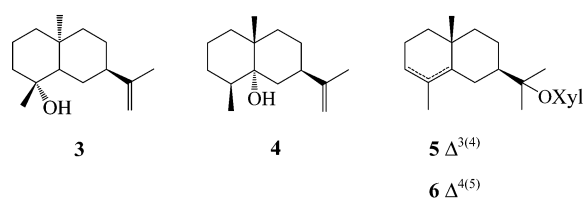
3.1.1 Eudesmanols. The novel eudesmanols **4–34** and their plant sources are detailed in Table 1. 4-Hydroxy-10-*epi*-eudesma-11-ene **3**²³ was synthesised from (–)-carvone, and the structure was established by X-ray analysis. The eudesmanols (\pm)-6-eudesmen-

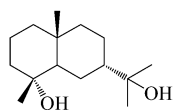
Table 1 Sources of eudesmanols

Sources	Compounds
<i>Artemisia annua</i>	29, 33, 34 ^{18,19}
<i>Calendula persica</i>	20, 21 ¹³
<i>Carthamus lanatus</i>	18, 19 ¹²
<i>Cassinia subtropica</i>	29, 32 ¹⁶
<i>Helichrysum italicum</i>	22 ¹⁴
<i>Iphiona mucronata</i>	23, 24, 26, 28 ¹⁵
<i>Iphiona scabra</i>	5, 6, 25, 27 ⁷
<i>Kleinia pendula</i>	4 ⁶
<i>Laggera alata</i>	10, 11 ¹⁰
<i>Laggera pterodonta</i>	9 ¹¹
<i>Ligularia veitchiana</i>	31 ^{20–22}
<i>Melampodium camphoratum</i>	12–17 ⁹
<i>Pegolettia oxydonta</i>	7, 8 ⁸
<i>Santolina rosmarinifolia</i>	30 ¹⁷

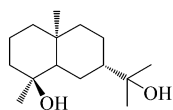
4 α -ol⁵ and (\pm)-selina-3,11-dien-9-ol²⁴ have also been the subject of synthetic studies, as has been the synthesis of α -dictyoptero²⁵ and platyllide.²⁶ A shorter route to the synthesis of (+)-junenol and isojunenol (together with their coumarate esters) starting from (–)-santonin has been developed, and (+)-junenol has been shown to be identical to 6-*epi*- β -verbesinol.²⁷

3.1.2 Eudesmandiols. New eudesmandiols **35–93** isolated from various plants are highlighted in Table 2. Kikkanol **B 88** showed inhibitory activity against rat lens aldose reductase.⁵³ The structure of teucdiol A was determined by X-ray analysis.⁵⁷ Eudesmane sesquiterpenes from the rhizomes of the Chinese

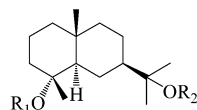




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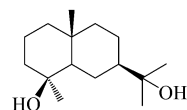


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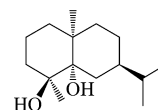


37 $R_1 = \text{Glc}$, $R_2 = \text{H}$

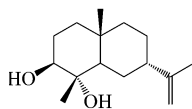
38 $R_1 = \text{H}$, $R_2 = \text{Glc}$



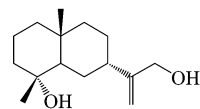
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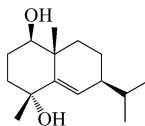
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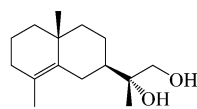
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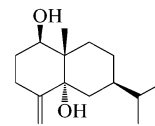
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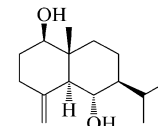
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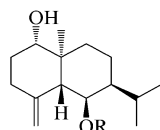
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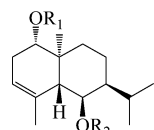


46



47 $R = \text{Ang}$

48 $R = \text{Cinn}$

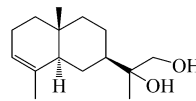


49 $R_1 = \text{Cinn}$, $R_2 = \text{H}$

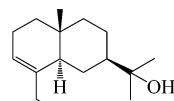
50 $R_1 = \text{H}$, $R_2 = \text{Cinn}$

51 $R_1 = \text{H}$, $R_2 = \text{Bz}(p\text{-OMe})$

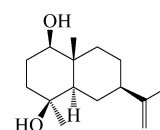
52 $R_1 = \text{H}$, $R_2 = \text{Glc}$



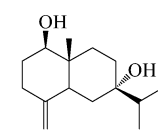
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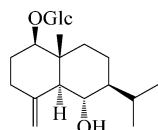
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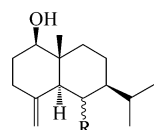
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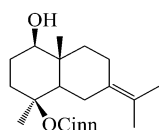


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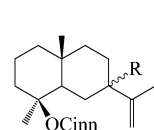


58 $R = \alpha\text{-OH}$

59 $R = \beta\text{-OH}$

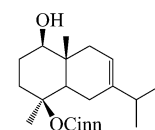


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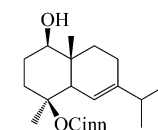


61 $R = \alpha\text{-OH}$

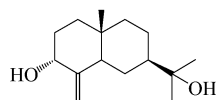
62 $R = \beta\text{-OH}$



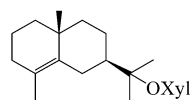
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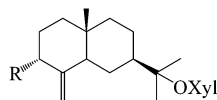
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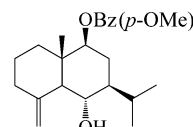
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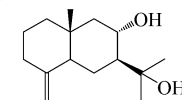
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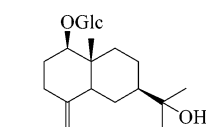
67 $R = \text{H}$
68 $R = \text{OH}$



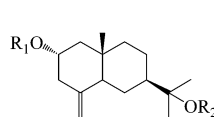
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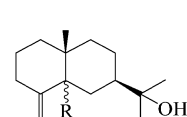


72 $R_1 = \text{H}$, $R_2 = \text{Glc}$

73 $R_1 = \text{Glc}$, $R_2 = \text{Glc}$

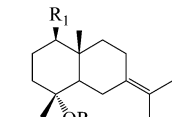
74 $R_1 = \text{Glc-Glc}$, $R_2 = \text{Glc}$

75 $R_1 = \text{Glc-Glc}$, $R_2 = \text{Glc-Glc}$



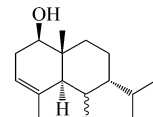
76 $R = \alpha\text{-OH}$

77 $R = \beta\text{-OH}$



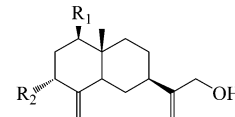
78 $R_1 = \text{OH}$, $R_2 = \text{Glc}$

79 $R_1 = \text{OGlc}$, $R_2 = \text{H}$



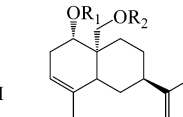
80 $R = \alpha\text{-OH}$

81 $R = \beta\text{-OH}$



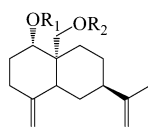
82 $R_1 = \text{OH}$, $R_2 = \text{H}$

83 $R_1 = \text{H}$, $R_2 = \text{OH}$



84 $R_1 = R_2 = \text{H}$

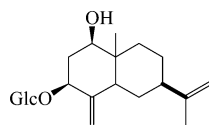
85 $R_1 = \text{H}$, $R_2 = \text{Ac}$



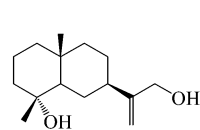
86 $R_1 = R_2 = \text{H}$

87 $R_1 = \text{H}$, $R_2 = \text{Ac}$

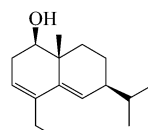
88 $R_1 = \text{Ac}$, $R_2 = \text{H}$



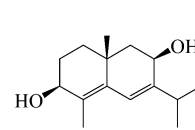
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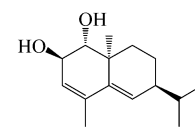
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Table 2 Sources of eudesmandiols

Sources	Compounds
<i>Ageratina glechonophylla</i>	58 ⁴¹
<i>Ambrosia artemisioides</i>	47–52 ³⁸
<i>Ambrosia elatior</i>	46, 69 ³⁷
<i>Artemisia arenaria</i>	43 ³⁵
<i>Artemisia eriopoda</i>	59 ⁴²
<i>Atractylodes lancea</i>	72–75 ⁴⁷
<i>Cassinia uncata</i>	53, 54 ¹⁶
<i>Chrysanthemum indicum</i>	65, 44, 91 ⁵³
<i>Erigeron annuus</i>	55–57 ³⁹
<i>Gonospermum fruticosum</i>	82, 83, 90 ⁵⁰
<i>Hypochoeris radicata</i>	89 ⁵²
<i>Iphiona scabra</i>	66, 67, 68 ⁷
<i>Isodon grandifolia</i>	35 ²⁹
<i>Jasonia candicans</i>	42 ³⁴
<i>Jasonia glutinosa</i>	44 ³⁶
<i>Jasonia montana</i>	76, 77 ⁴⁸
<i>Laggera alata</i>	70 ⁴⁵
<i>Laggera crispata</i>	41 ³³
<i>Laggera pterodonta</i>	36–38, 78, 79 ^{30,31}
<i>Ligularia duciformis</i>	92 ^{54,55}
<i>Ligularia hodgsonii</i>	45 ²¹
<i>Pluchea arguta</i>	35, 39 ^{28,32}
<i>Pluchea dioscoridis</i>	80, 81 ⁴⁹
<i>Pluchea indica</i>	71 ⁴⁶
<i>Polyachirus sphaerocephalus</i>	84–88 ⁵¹
<i>Santolina insularis</i>	93 ⁵⁶
<i>Senecio microglossus</i>	58 ⁴⁰
<i>Ursinia trifida</i>	39, 40 ³
<i>Verbesina oerstediana</i>	60–64 ⁴³

herb *Atractylodes ovata* have been shown to have cytotoxic activity against leukaemia cell lines.⁵⁸ The microbiological transformation of ilicic acid and kudtiol by cultures of *Cunninghamella echinulata* has been studied.^{59,60} The microbiological transformations of 4 α - and 4 β -hydroxyeudesmane derivatives by the filamentous fungus *Gliocladium roseum* have also been reported.⁶¹

A general enantiospecific approach to the synthesis of polyfunctional eudesmanes based on ring-closing metathesis has been reported.⁶² (+)-Dihydrocarvone has been used as a starting point for the syntheses of (–)-10-*epi*-5 β ,11-dihydroxyeudesmane and (–)-4,10-*epi*-5 β ,11-dihydroxyeudesmane.⁶³ The first total synthesis of both C-11 epimers of 13-hydroxy- α -eudesmol has been completed. This work has permitted the absolute configuration of the natural diol to be established as **53**, and also the preparation of (+)- α -selinene to be accomplished.⁶⁴ The first enantioselective total syntheses of (+)-chrysanthemol,⁶⁵ (+)-selina-3,11-dien-9-ol⁶⁶ and (–)-rishitin⁶⁷ have been described, and racemic preparations of dehydrochamaecyneaol⁶⁸ and balanitol have been reported.⁶⁹ α -Eudesmaol,⁷⁰ occidol⁷¹ and tavaopallescensin⁷² have been the subject of total synthesis, and a formal total synthesis of (–)-dehydrochamaecyneaol has been presented.⁷³ Eudesman-4-en-1 β ,11-diol has been synthesised as its racemate.⁷⁴

3.1.3 Eudesmantriols. Recently isolated eudesmantriols **94–138** and their plant sources are listed in Table 3. The structure of mucrolidin **104** was determined by X-ray analysis.⁷⁹ Kikkanol **A 118** showed inhibitory activity against rat lens aldose reductase.⁵³ The microbiological transformation of β -selinene by the plant pathogenic fungus *Glomerella cingulata* has been studied.⁹¹ Enantioselective total syntheses of kudtriol, 5-*epi*-kudtriol and its C-11 epimer^{92–94} have been accomplished.

Table 3 Sources of eudesmantriols

Sources	Compounds
<i>Achillea clypeolata</i>	129 ⁸⁷
<i>Ainsliaea cordifolia</i>	124, 125 ⁸²
<i>Artemisia eriopoda</i>	96 ⁴²
<i>Artemisia monosperma</i>	132 ⁸⁸
<i>Atractylodes lancea</i>	114, 115, 131 ^{47,86}
<i>Chrysanthemum indicum</i>	118 ⁵³
<i>Erigeron brevicarpus</i>	127 ⁸⁴
<i>Iva frutescens</i>	103, 104 ⁷⁸
<i>Jasonia glutinosa</i>	108 ³⁶
<i>Laggera alata</i>	130 ⁸⁵
<i>Laggera pterodonta</i>	97–100, ^{30,31} 105, 106, ¹¹ 109–112 ⁸⁰
<i>Pallenis spinosa</i>	121, 122 ⁸¹
<i>Pluchea indica</i>	113, 123 ⁴⁶
<i>Polyachirus fuscus</i>	135–138 ⁹⁰
<i>Rhaponticum uniflorum</i>	134 ⁸⁹
<i>Santolina chamaecyparissus</i>	126 ⁸³
<i>Santolina insularis</i>	133 ⁵⁶
<i>Saussurea laniceps</i>	94 ⁷⁵
<i>Santolina rosmarinifolia</i>	119 ¹⁷
<i>Tanacetopsis mucronata</i>	107 ⁷⁹
<i>Tessaria integrifolia</i>	101, 102 ⁷⁷
<i>Verbesina eggersii</i>	95 ⁹⁷
<i>Verbesina oerstediana</i>	116, 120, 128 ⁴³
<i>Verbesina virgata</i>	116, 117 ⁷⁶

3.1.4 Polyhydroxylated eudesmanes. 3,4,7,8-Tetrahydroxy eudesmanes **139** and **140** have been isolated from the methanolic extract of *Tessaria integrifolia*.⁷⁷ The tetrahydroxy eudesmanes **141**, **142** and **143** have also been found in the Asteraceaeous plants *Ainsliaea cordifolia*,⁸² *Senecio flammeus*,^{95,96} and *Verbesina eggersii*,⁹⁷ respectively. The pentahydroxy eudesmane **144** was isolated from *Calendula arvensis*.⁹⁸

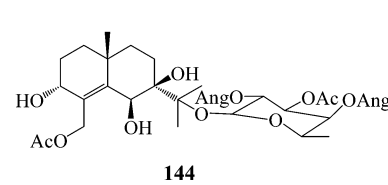
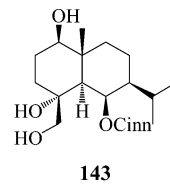
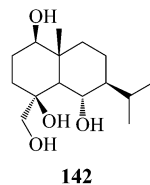
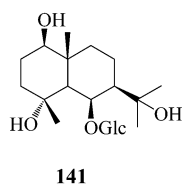
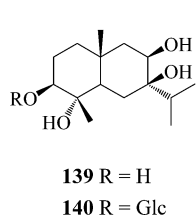
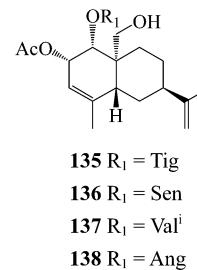
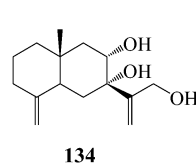
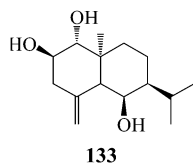
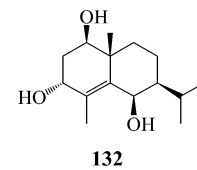
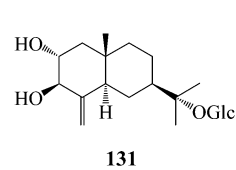
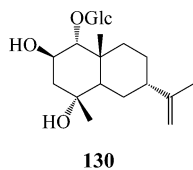
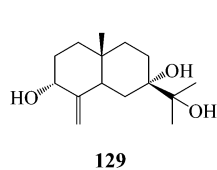
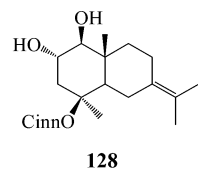
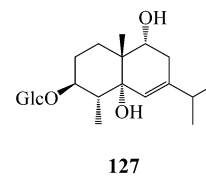
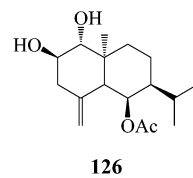
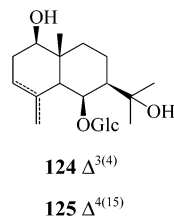
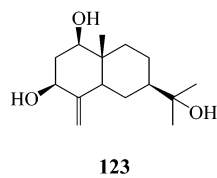
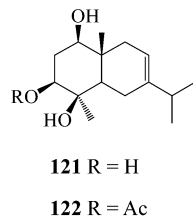
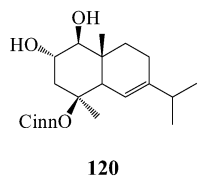
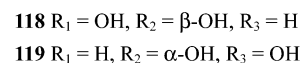
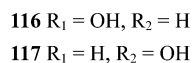
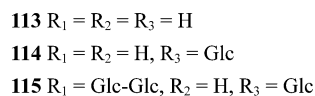
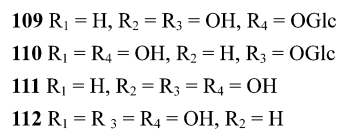
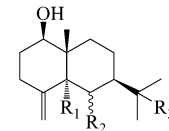
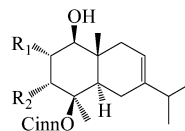
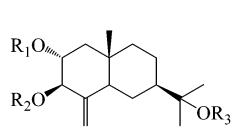
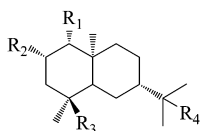
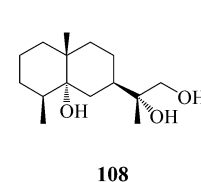
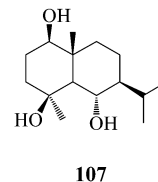
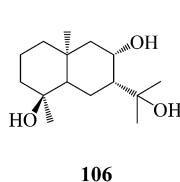
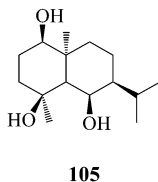
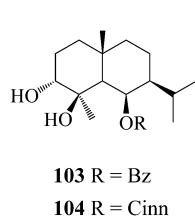
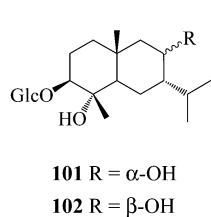
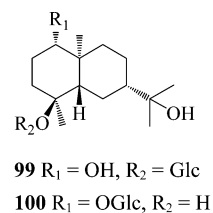
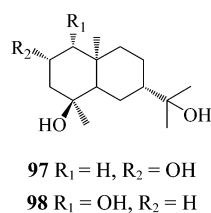
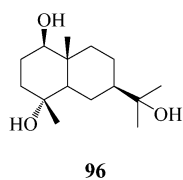
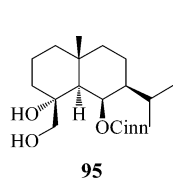
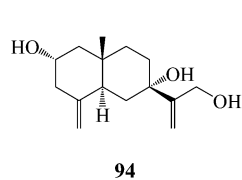
Two novel polyhydroxyl 7-*epi*-eudesmane derivatives have been isolated from the microbial transformation of a (1 α ,10 β),(4 β ,5 α)-diepoxygermacrane using the hydroxylating fungus *Rhizopus nigricans*. The absolute configurations of these compounds was confirmed by single-crystal X-ray analyses.⁹⁹ The 1 α - and 2 α -hydroxyselinane derivatives have been prepared by chemical semi-synthesis and biotransformation with *Rhizopus nigricans*, using vulgarin as starting material.¹⁰⁰

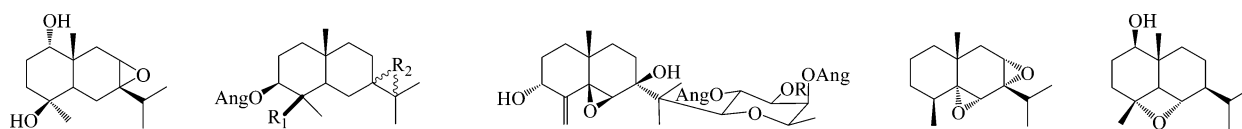
3.2 Eudesmane epoxides

The eudesmane epoxides **146–171** and their corresponding plant sources are indicated in Table 4. The structure of epoxyplodiol **145** has been determined by single crystal X-ray diffraction

Table 4 Sources of eudesmane epoxides

Sources	Compounds
<i>Ambrosia artemisioides</i>	154, 155 ³⁸
<i>Atractylodes japonica</i>	161 ¹⁰⁷
<i>Baccharis dracunculifolia</i>	153 ¹⁰³
<i>Calendula arvensis</i>	148, 149, 150 ⁹⁸
<i>Chrysanthemum indicum</i>	159 ¹⁰⁶
<i>Curculigo capitulata</i>	152 ¹⁰⁴
<i>Epaltes gariepina</i>	164–168 ⁴⁵
<i>Erigeron philadelphicus</i>	156 ¹⁰⁵
<i>Ixeris repens</i>	162, 163 ¹⁰⁸
<i>Liabum floribundum</i>	151 ¹⁰²
<i>Ondetia linearis</i>	169, 170 ¹⁰⁹
<i>Pluchea quitoc</i>	146, 147, ¹⁰¹ 171 ¹¹⁰
<i>Polyachirus sphaerocephalus</i>	157, 158 ⁵¹





145

146 R₁ = α -OH, R₂ = β -epoxy147 R₁ = β -OAc, R₂ = α -epoxy

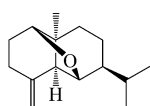
148 R = Ac

149 R = C(O)Prⁱ

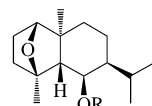
150 R = Mebu

151

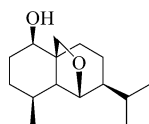
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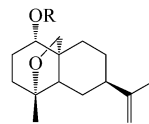
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154 R = Cinn

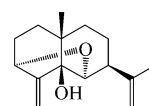
155 R = COC₆H₄OMe

156

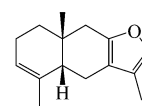


157 R = H

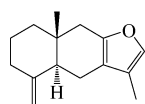
158 R = Ac



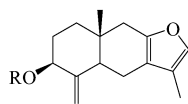
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160

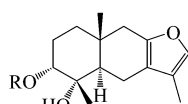


161



162 R = H

163 R = Ac

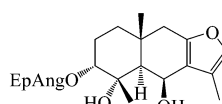


164 R = H

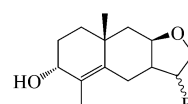
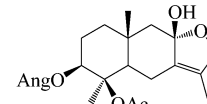
165 R = EpAng

166 R = Valⁱ

167 R = Ang



168

169 R = α -Me170 R = β -Me

171

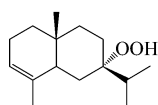
analysis.¹¹¹ The furanosesquiterpene atractylon showed moderate, but selective, 5-lipoxygenase inhibitory activity.¹¹² (+)-Dihydrocarvone has been used as a starting material for the first syntheses of (+)-5 α -hydroxy- β -selinene and (-)-5 β -hydroxy- β -selinene.¹¹³ The synthesis of (+)-1,2-dihydrotubipofuran **160**, starting from α -santonin, has been completed.¹¹⁴ Synthetic studies towards the furosesquiterpenes have been developed,¹¹⁵ and a synthesis of both enantiomers of acetoxytubipofuran has been reported.¹¹⁶

3.3 Eudesmane peroxides

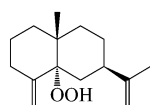
Novel peroxide-substituted eudesmanes **172–187** and their corresponding plant sources are detailed in Table 5. There is very

Table 5 Sources of eudesmane peroxides

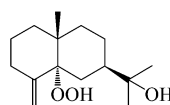
Sources	Compounds
<i>Achillea clypeolata</i>	184 ¹²¹
<i>Ambrosia artemisioides</i>	180–182 ²⁸
<i>Artemisia annua</i>	173 ¹⁹
<i>Artemisia douglasiana</i>	174 ¹¹⁸
<i>Aster oharai</i>	172 ¹¹⁷
<i>Brontonia discodea</i>	183, 185 ¹²⁰
<i>Calea prunifolia</i>	177 ¹¹⁹
<i>Dittrichia viscosa</i>	187 ¹²³
<i>Isocoma coronopifolia</i>	186 ¹²²
<i>Verbesina oerstediana</i>	175, 176, 178, 179 ⁴³



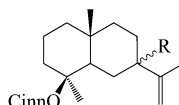
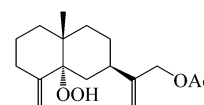
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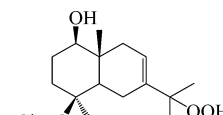
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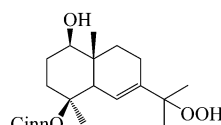
174

175 R = α -OOH176 R = β -OOH

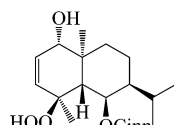
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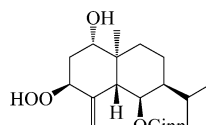
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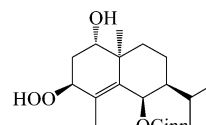
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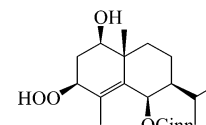
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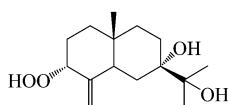
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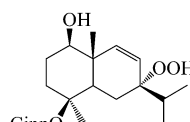
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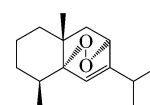
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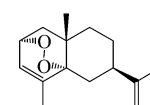
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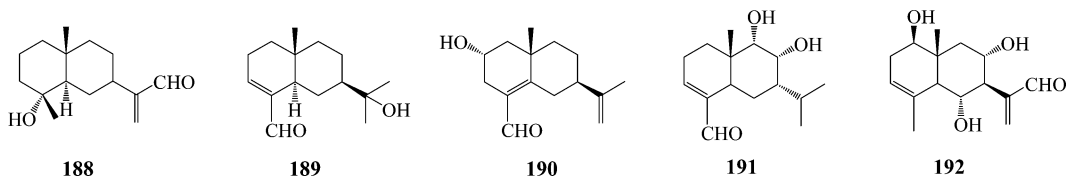
185



186



187



little literature on the bioactivity or synthesis of these structurally interesting eudesmane sesquiterpenoids.

3.4 Eudesmane aldehydes

The eudesmane aldehyde **188** has been found in *Vernonia glabra* var. *glabra*.¹²⁴ The other α,β -unsaturated aldehydes **189**, **190**, **191** and **192** have been identified in *Cassinia uncata*,¹⁶ *Tetragonotheca ludoviciana*,¹²⁵ *Chrysanthemum indicum*,⁵³ and *Tanacetopsis mucronata*,¹²⁶ respectively. The inhibitory activity of compound **191** against rat lens aldose reductase has been studied.⁵³ A series of sesquiterpene compounds possessing eudesmane carbon skeletons were tested as gastric cytoprotective agents on male Wistar rats. The presence of an α,β -unsaturated aldehyde on the C-7 side chain together with a hydroxyl group at C-4 appears to be a prerequisite for their observed anti-ulcerogenic activity. In an attempt to establish new molecular structural requirements for this gastric cytoprotective activity, a structure–activity study has been performed.¹²⁷

A new salen–manganese(III) complex bearing a sesquiterpene salicylaldehyde derivative has been used in the catalytic epoxidation of unfunctionalised olefins, with iodosylbenzene and molecular oxygen–pivalaldehyde as the terminal oxidant.¹²⁸

3.5 Eudesmane ketones

A series of recently isolated eudesmane ketones **193–272** and their plant sources are highlighted in Table 6. The novel sesquiterpene ketone **194** was the isomer from the genus *Nephtia*.¹³⁰ The new cauthemone derivatives **212–218**, **220** and **221** have been identified from plants of the genus *Tessaria*, while **237**, **240**, **241**, **243** and **250** have been found in *Pluchea symphytifolia*, indicating that there is a phytochemical relationship between these two Asteraceae genera. The structure of 5-*O*-acetylcauhtemomyl-6-*O*-2,3'-epoxy-2-methylbutyrate, isolated from *Pluchea carolinensis*, has been determined by X-ray analysis.¹⁶³ Sesquiterpene **250**, which was isolated from *Pluchea suaveolens*¹⁶⁴ and *Epaltes brasiliensis*,¹⁶⁵ has been revised from the 11-hydroxy- to the 11-peroxy-derivative.¹⁴² The structure of the sesquiterpene ketone carissone has been confirmed by X-ray analysis,¹⁶⁶ and the absolute configuration of cauhtemone has been revised to **210**.¹⁶⁷

The antibacterial activities of **246**¹⁵² and **263**¹⁵⁹ have been studied. The antiprotozoal activity of a range of known sesquiterpenes from *Jasonia glutinosa* has been explored and the antibacterial activities of carissone, dehydrocarissone and carindone have been evaluated.¹⁶⁸

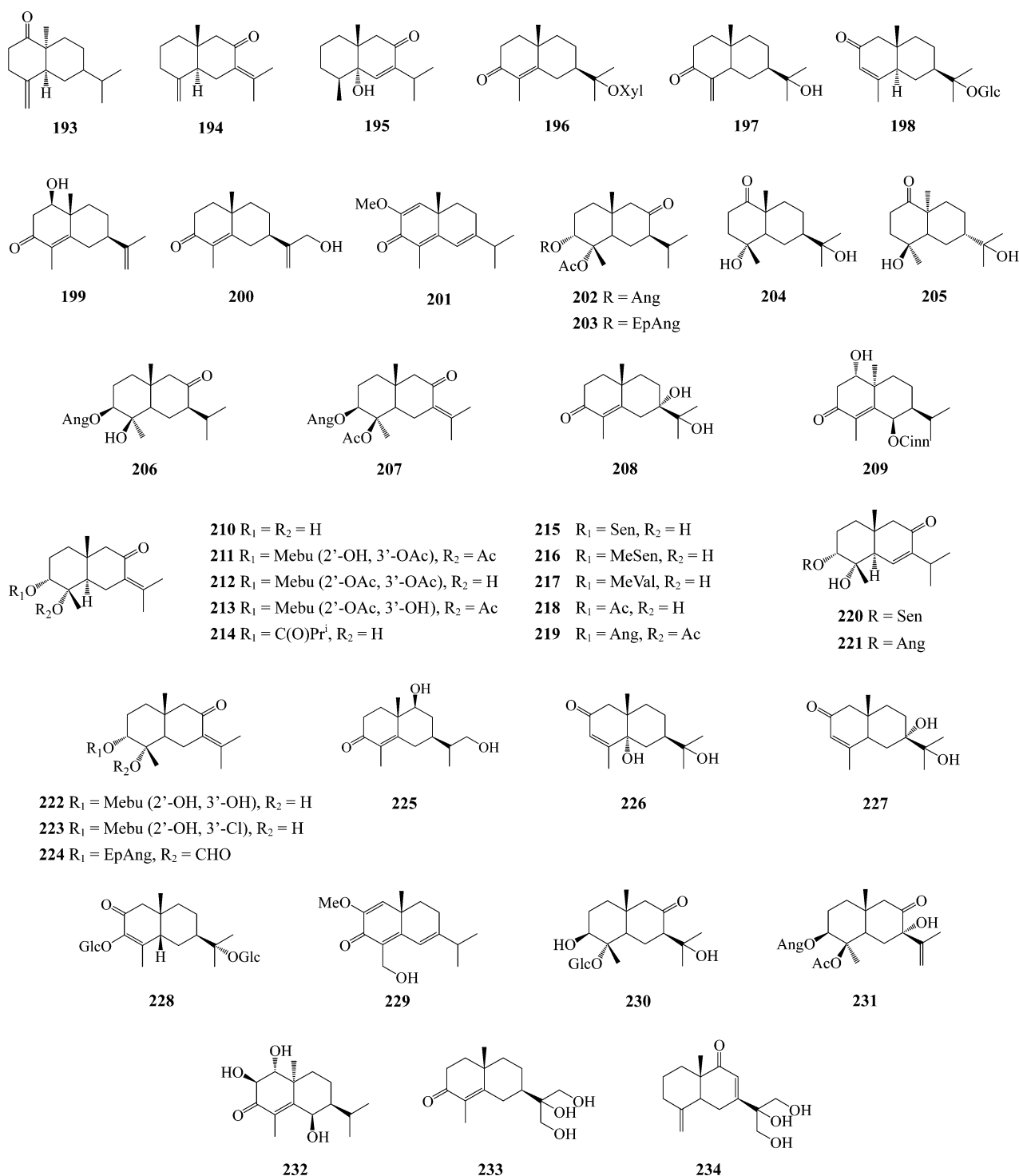
The biotransformation of 4 β -hydroxyeudesmane-1,6-dione by the fungi *Gliocladium roseum* and *Exserohilum halodes* has been studied.¹⁶⁹ Alantolactone has been shown to induce apoptosis in jurkat leukaemia T cells.¹⁷⁰ A chemical–microbiological synthesis of 6 β -eudesmanolides has been achieved with the fungus *Rhizopus nigricans*.¹⁷¹ The microbiological transformation of

Table 6 Sources of eudesmane ketones

Sources	Compounds
<i>Achillea clypeolata</i>	208 , 227 ¹²¹
<i>Achillea holosericea</i>	233 ¹⁵⁵
<i>Ambrosia artemisioides</i>	209 ³⁸
<i>Artemisia afra</i>	200 ¹³⁶
<i>Artemisia caerulescens</i>	199 , 268 ^{133,134}
<i>Artemisia eriopoda</i>	204 , 226 ⁴²
<i>Artemisia herba-alba</i>	269 ¹⁶¹
<i>Atractylodes japonica</i>	194 ¹⁰⁷
<i>Atractylodes lancea</i>	198 , ⁸⁶ 228 ^{47,86}
<i>Baccharis boliviensis</i>	271 ¹⁶²
<i>Blumea alata</i>	210 , 211 , 255–258 , 264 , 265 ¹³⁹
<i>Cassinia uncata</i>	225 , 270 ¹⁶
<i>Epaltes divaricata</i>	219 ¹⁴³
<i>Epaltes mexicana</i>	222 , ^{144,145} 241 , ¹⁴⁴ 263 ¹⁵⁹
<i>Hypochoeris radicata</i>	199 ¹³⁵
<i>Iphionea scabra</i>	196 ⁷
<i>Laggera alata</i>	224 ⁴⁵
<i>Laggera crispata</i>	235 ³³
<i>Laggera pterodonta</i>	205 , 226 ¹¹
<i>Liabum floribundum</i>	195 ¹⁰²
<i>Parthenium argentatum</i>	201 , 229 , 272 ¹³⁷
<i>Pluchea arguta</i>	213 , ¹⁴¹ 236 , ¹⁴⁷ 238 , ¹⁴⁵ 239 , ¹⁴⁹ 245 , 246 , ^{151,152} 248 , ³² 250 , ¹⁵⁰ 252 , ¹⁵³ 253 , ¹⁵⁴ 254 , ¹⁵¹ 260–262 , ^{147,151,158} 266 , 267 ^{160,158}
<i>Pluchea carolonesis</i>	223 ¹⁴⁶
<i>Pluchea indica</i>	212 ¹⁴⁰
<i>Pluchea odorata</i>	237 , ¹⁴⁸ 242 , 244 , 247 , 251 ¹⁵⁰
<i>Pluchea quitoc</i>	243 , 249 , ¹¹⁰ 231 , 259 , ¹⁰¹ 206 , 207 ^{101,110}
<i>Pluchea symphytifolia</i>	241 , 243 , 250 ¹⁴²
<i>Santolina insularis</i>	232 ⁵⁶
<i>Saussurea lappa</i>	230 ⁴⁵
<i>Saussurea parviflora</i>	234 ^{156,157}
<i>Senecio bracteolatus</i>	199 ¹³²
<i>Solidago gigantea</i>	193 ¹²⁹
<i>Sphaeranthus indicus</i>	197 ¹³¹
<i>Tessaria integrifolia</i>	202 , 203 , ¹³⁸ 214–218 , 220 , 221 , 240 ¹⁴²

eudesmane derivatives by the fungus *Curvularia lunata* has also been reported.¹⁷² The atractylon content of clonally propagated plants of *Atractylodes japonica* has been determined¹⁰⁷ and the allelochemical effects of eudesmane sesquiterpenes on *Tribolium castaneum* larvae have been investigated.¹⁷³

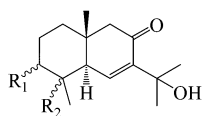
A preparation of the HS-toxin A aglycone, starting from (–)-carvone, has been described.¹⁷⁴ (+)-Carvone has been used as the starting material in the synthesis of (+)- α -cyperone,¹⁷⁵ and the first total synthesis of (+)-5 α -hydroxyisopterocarpolone, starting from (+)-dihydrocarvone, has been described.¹⁷⁶ The hydroxylation of sesquiterpenes by enzymes from chicory roots has been investigated.¹⁷⁷ 3 β -Angeloyloxy-4 β -hydroxyeudesman-8-one **206** has been synthesised by an unambiguous route,¹⁷⁸ but its spectroscopic data was different from that of the natural product, to which this structure had been assigned.^{101,179} An efficient and stereoselective synthesis of (+)- α -cyperone has been devised,¹⁸⁰ and the eudesmanolide artemisin has been used as a starting material in the preparation of 9-oxy-functionalised natural eudesmanes.¹⁸¹ The total syntheses of (\pm)-cauhtemone¹⁸²



and (±)-eudesma-5,7(11)-dien-8-one has been reported.¹⁸³ The chemical transformation of α -santonin into two 8-oxo- β -cyperone derivatives has been reported.¹⁸⁴ A shorter, more efficient method for the preparation of 11,12-dihydroxyeudesmanolides, starting from α -santonin, has been reported,¹⁸⁵ and a total synthesis of the naphthofurandione maturinone has been achieved.¹⁸⁶ Phytuberin has been synthesised starting from carvone.¹⁸⁷ A facile synthesis of 3-oxo-7 α H-eudesma-4 β ,12-diol, previously isolated from *Casima uncata*, has been described.¹⁸⁸ An enantioselective synthesis of selina-1,3,7(11)-trien-8-one has been accomplished.¹⁸⁹

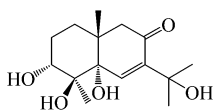
3.6 Eudesmane carboxylic acids

Recently isolated eudesmane carboxylic acids **273–423** from a number of different plants are listed in Table 7. The structure of vachanic acid²⁵⁸ and **411**²⁵⁹ have been conformed by X-ray studies. X-Ray crystallographic studies have also revealed catameric carboxyl-to-ketone hydrogen bonding in the bicyclic sesquiterpene keto acid **406**.²⁴² The aerial parts of *Flourensia thurifera* contain 3 β -hydroxycostic acid **283** and a mixture of two acyl derivatives **284** and **285**. The toxicity of these substances toward *Artemia salina*

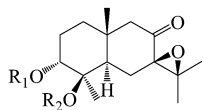


- 235** $R_1 = \alpha\text{-OMebu (2'-OH, 3'-OH)}$, $R_2 = \alpha\text{-OH}$
236 $R_1 = \alpha\text{-OMebu (2'-OH, 3'-OH)}$, $R_2 = \beta\text{-OH}$
237 $R_1 = \beta\text{-OMebu (2'-OH, 3'-OH)}$, $R_2 = \alpha\text{-OH}$
238 $R_1 = \beta\text{-OMebu (2'-OH, 3'-Cl)}$, $R_2 = \alpha\text{-OH}$
239 $R_1 = \beta\text{-OH}$, $R_2 = \alpha\text{-OH}$
240 $R_1 = \alpha\text{-OSen}$, $R_2 = \alpha\text{-OH}$
241 $R_1 = \alpha\text{-OAng}$, $R_2 = \alpha\text{-OH}$
242 $R_1 = \alpha\text{-OAng}$, $R_2 = \alpha\text{-OAc}$
243 $R_1 = \beta\text{-OAng}$, $R_2 = \alpha\text{-OH}$

- 244** $R_1 = \alpha\text{-OMebu (2'-OH, 3'-Cl)}$, $R_2 = \alpha\text{-OAc}$
245 $R_1 = \alpha\text{-OMebu (2'-OH, 3'-Cl)}$, $R_2 = \beta\text{-OAc}$
246 $R_1 = \alpha\text{-OMebu (2'-OH, 3'-Cl)}$, $R_2 = \alpha\text{-OAc}$
247 $R_1 = \alpha\text{-OMebu (2'-OH, 3'-Cl)}$, $R_2 = \alpha\text{-OH}$
248 $R_1 = \alpha\text{-OMebu (2'-OH, 3'-OH)}$, $R_2 = \alpha\text{-OAc}$
249 $R_1 = \beta\text{-OAng}$, $R_2 = \beta\text{-OAc}$
250 $R_1 = \alpha\text{-OEpAng}$, $R_2 = \alpha\text{-OH}$
251 $R_1 = \alpha\text{-OEpAng}$, $R_2 = \alpha\text{-OAc}$
252 $R_1 = \alpha\text{-OEpAng}$, $R_2 = \beta\text{-OAc}$
253 $R_1 = \beta\text{-OMebu (2'-OH, 3'-Cl)}$, $R_2 = \beta\text{-OAc}$



254

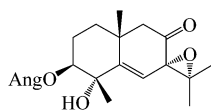


255 $R_1 = \text{Ang}$, $R_2 = \text{H}$

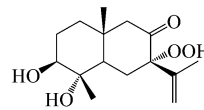
256 $R_1 = \text{Ang}$, $R_2 = \text{Ac}$

257 $R_1 = \text{EpAng}$, $R_2 = \text{H}$

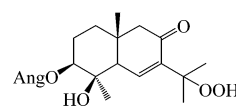
258 $R_1 = \text{EpAng}$, $R_2 = \text{Ac}$



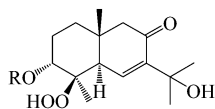
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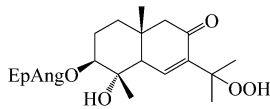
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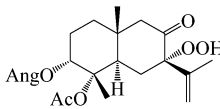
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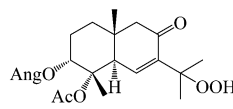
262 $R = \text{Mebu (2'-OH, 3'-Cl)}$



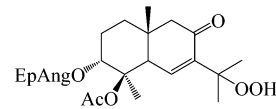
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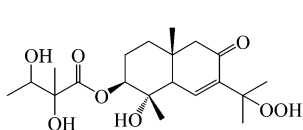
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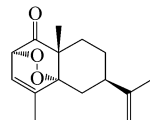
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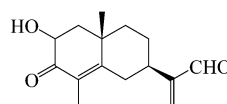
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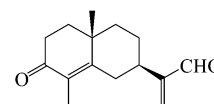
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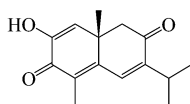
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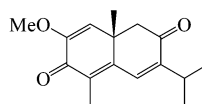
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270



271



272

and their antifeedant properties against *Spodoptera littoralis* have been evaluated.¹⁹⁹ Compound **411** exhibited potent antimicrobial activity against the six bacteria *Staphylococcus aureus*, *Bacillus subtilis*, *Micrococcus luteus*, *Escherichia coli*, *Bacillus cereus* and *Salmonella enteritides*.²⁶⁰ The sesquiterpene 12-carboxyeudesma-3,11(13)-diene and the xanthnolide tomentosin were the major compounds responsible for the ichthyotoxicity shown by *Dittrichia graveolens*.²⁶¹ A mechanistic approach to study the *in vivo* anti-inflammatory activity of various sesquiterpenes isolated from *Inula viscosa* has been carried out.²⁶² The anti-inflammatory properties of other sesquiterpenes from *Jasonia glutinosa*²⁶³ have also been tested.

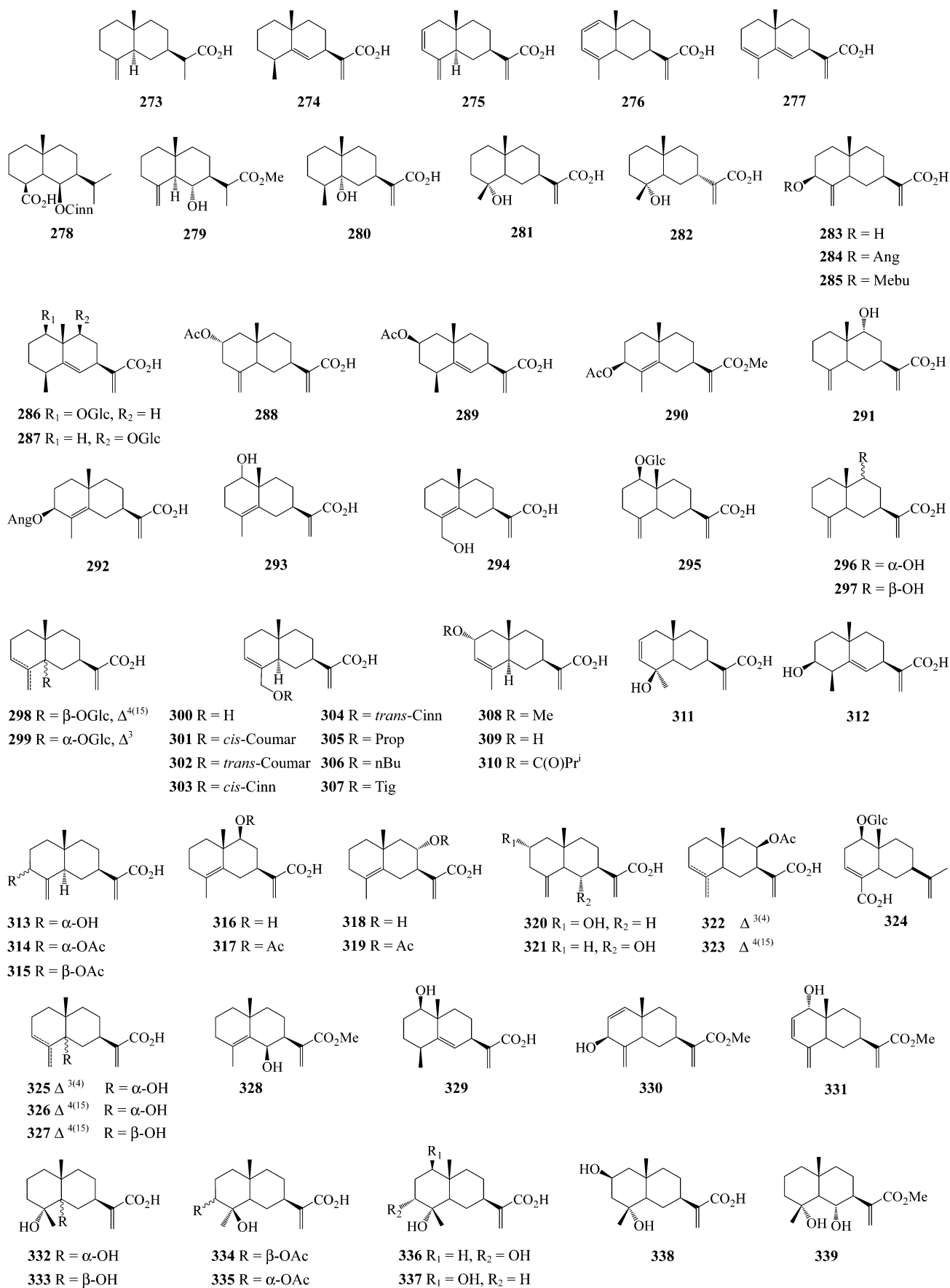
The bioconversion of several eudesmane derivatives using the fungi *Rhizopus nigricans* and *Curvularia lunata* has been studied.^{264,265} The microbiological transformation of the eudesmane sesquiterpenes ilicic acid and plectranthone by the fungi *Aspergillus niger*²⁶⁶ and *Beauveria bassiana*,²⁶⁷ respectively, have been described, and the microbiological transformation of ilicic

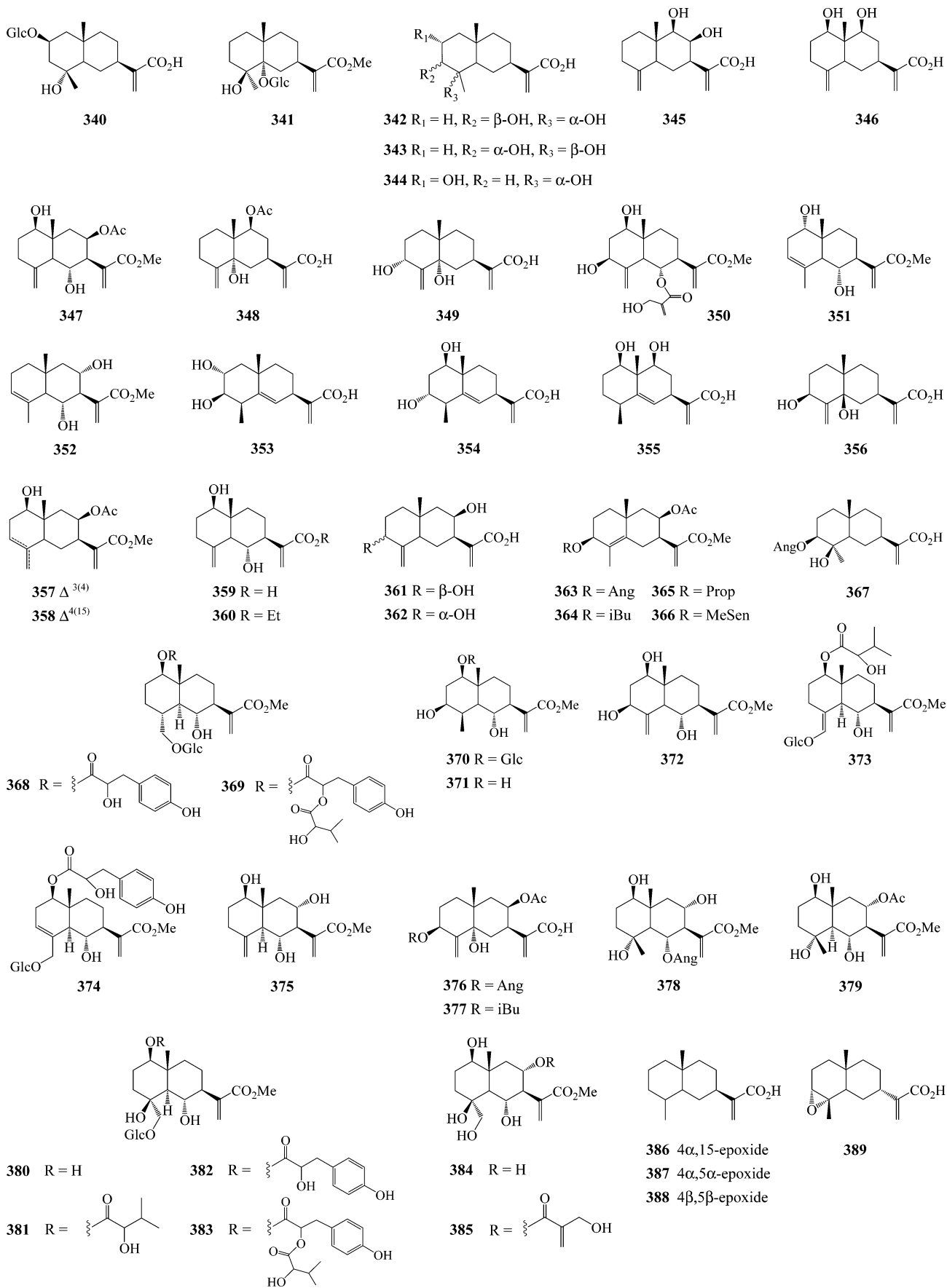
acid by cultures of *Cunninghamella echinulata* has also been evaluated.^{59,60}

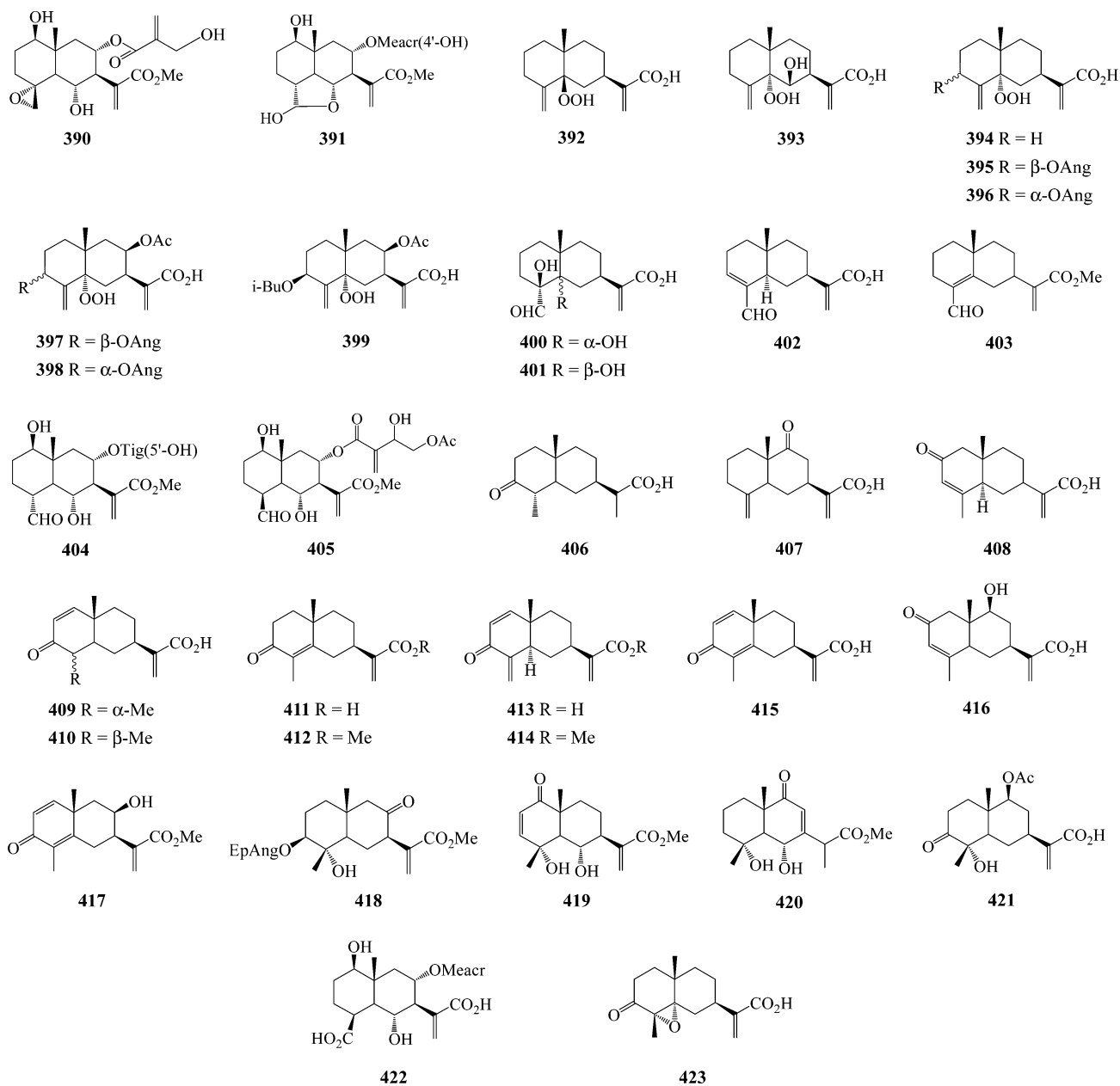
(±)-Dihydrocarvone has been used as the starting material in the synthesis of (+)-3-oxoeudesma-4,11(13)-dien-12-oic acid,²⁶⁸ and santonin has been employed in the preparation of furanoeudesma-1,3-diene and tubipofurane.²⁶⁹ The total synthesis of the eudesmanic acid **415**, starting from (+)-dihydrocarvone, has been achieved.^{249,250} An enantioselective total synthesis of eudesma-3,11(13)-dien-12-oic acid.²⁷⁰ has also been completed. The recombinant sesquiterpene synthases from grand fir δ -selinene synthase produces more than 30 sesquiterpene olefins from the acyclic precursor farnesyl diphosphate.²⁷¹

3.7 Eudesmanolides

3.7.1 Eudesmane-12,6-olides. A wide range of structurally diverse eudesmane-12,6-olides have been reported from the Asteraceae family, and a good deal of biological activity,







biotransformation and chemical syntheses studies about those lactones have been completed.

The lactones **424–742** and their corresponding plant sources are outlined in Table 8. Lasolide **506** and isolasolide **507** have been obtained from plants of the genus *Lasera*, and the structure of **506** has been revised.³¹⁴ The structures of *7 α* -hydroxycostunolide³³⁸ badkhisinin **443** and oopodin **660** have been determined by X-ray analysis,³⁹⁵ and the stereochemistry of tauremisin **707** has been determined by single-crystal X-ray diffraction analysis.²⁰¹ The structure of torrentin, an eudesmanolide isolated from *Artemisia herba-alba*, has been revised to **487**.³⁹⁹ The lactone **432**, also obtained from *Artemisia herba-alba*,²⁷⁹ has had its structure revised, as the original configuration assigned at C-11 was erroneous.²⁷⁸ Conformational analyses of *6 α* - and *6 β* -eudesmanolides, as well as *8 α* - and *8 β* -eudesmanolides, have been carried out.³⁹⁶ The structure of 1-*O*-acetylbritannilactone

has been determined by X-ray analysis,³⁹⁷ whilst the absolute configuration of glaucescenolide has been confirmed by chemical synthesis.³⁹⁸ The known eudesmanolide magnolialide has been obtained from *Cichorium intybus* and shown to be identical to cichoriolide (another eudesmanolide), and cichopumulide (a guaianolide). Consequently, the structures of these last two lactones must be corrected. Moreover, the structures of their natural 11 β ,13-dihydro derivatives and the glycosides cichorioside and sonchuside C should also be amended.³⁹⁹ The structures of the alkaline reduction products of santonin have been determined.⁴⁰⁰ A straightforward procedure for the introduction of a double bond between C-7 and C-11 in the eudesmanolide skeleton has been described.⁴⁰¹

Sphaeranthanolide **498** exhibited immunostimulant activity in the jerne plaque assay.⁴⁰² The toxicity and antifeedant activity of several eudesmanolides from two *Encelia* species against larvae of

Table 7 Sources of eudesmane carboxylic acids

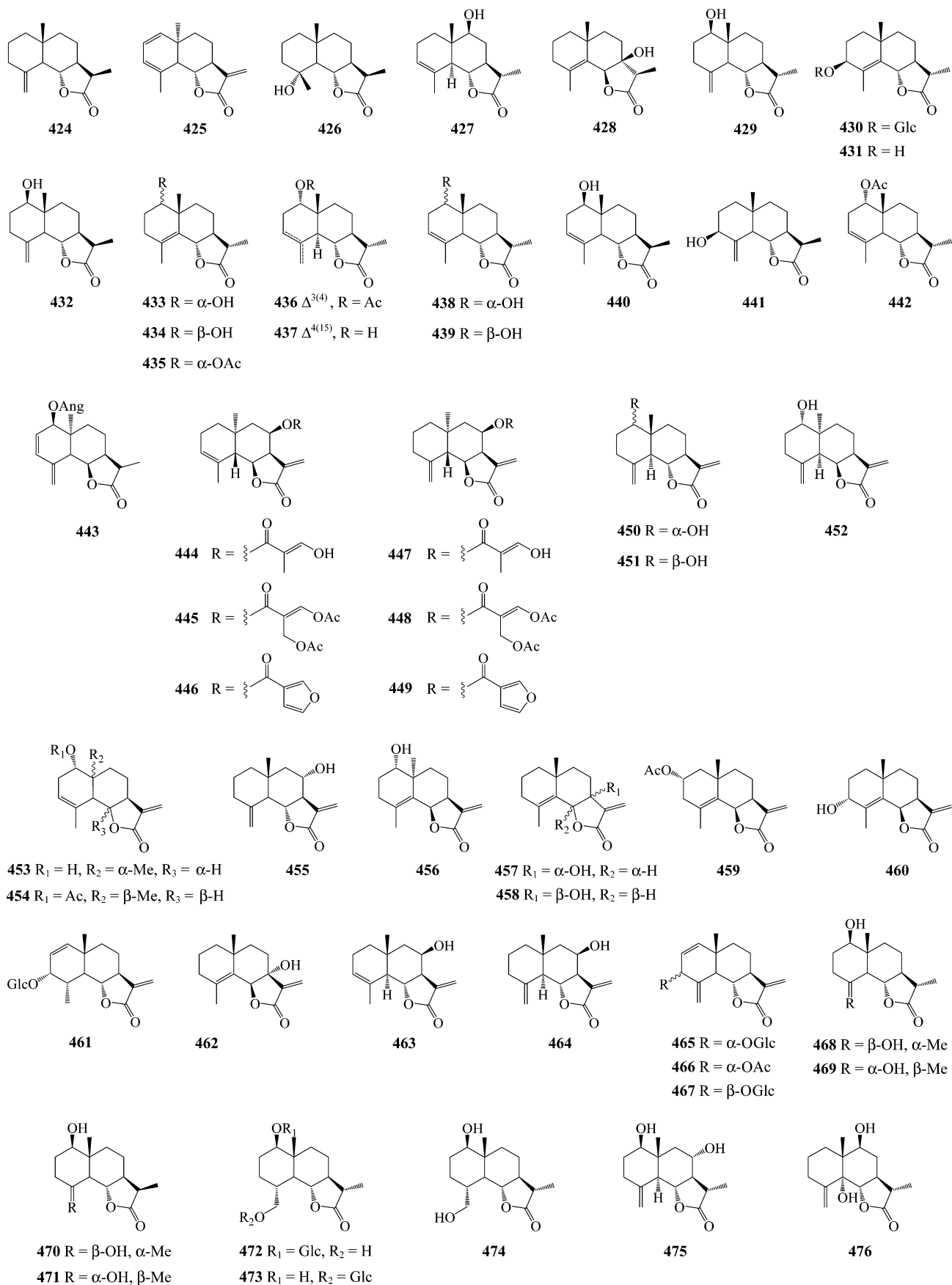
Sources	Compounds
<i>Achillea schischkinii</i>	420 ²⁵⁴
<i>Ambrosia elatior</i>	293 ³⁷
<i>Apalochlamys spectabilis</i>	325–327, ^{48,197} 280, 386–388 ¹⁹⁷
<i>Arctotis aspera</i>	413 ²⁴⁷
<i>Artemisia altaiensis</i>	412 ²⁴⁶
<i>Artemisia</i> genus	406 ²⁴²
<i>Artemisia herba-alba</i>	281, ¹⁹⁸ 326 ²¹⁸
<i>Artemisia mongolica</i>	352 ²²⁹
<i>Artemisia pectinata</i>	318, 319 ²¹³
<i>Artemisia phaeolepis</i>	421 ²⁵⁵
<i>Artemisia rutifolia</i>	350, ²²⁷ 372 ²³⁶
<i>Artemisia tournefortiana</i>	316, ²¹¹ 316, 317, ²¹² 348, ³¹ 407 ²¹¹
<i>Artemisia vulgaris</i>	331, 415 ²²⁰
<i>Brocchia cinerea</i>	378 ²³⁷
<i>Cassinia uncata</i>	283, 290, 300–307, 375, 379, 402 ¹⁶
<i>Centaurea arguta</i>	330, 414 ²¹⁹
<i>Centaurea attica</i>	405 ²⁴¹
<i>Centaurea chilensis</i>	291 ²⁰¹
<i>Centaurea tweediei</i>	422 ²⁵⁷
<i>Cratystylis conocephala</i>	320 ²¹⁵
<i>Dittrichia viscosa</i>	275, ¹⁹² 309, 313 ²⁰⁷
<i>Eriocephalus pauperrimus</i>	292, 361–367, 376, 377, 392, 394–399, 423 ²⁰²
<i>Eupatorium quadrangulare</i>	279 ¹⁹⁶
<i>Ferreyranthus fruticosus</i>	417 ²⁵¹
<i>Flourensia macrophylla</i>	296, 297, 345, 346, 407 ²⁰⁴
<i>Flourensia thurifera</i>	283–285 ¹⁹⁹
<i>Geigeria rigida</i>	322, 323 ²¹⁷
<i>Haeckeria punctulata</i>	314, 315, ²¹⁰ 334, 335 ²²³
<i>Helianthus annuus</i>	276 ¹⁹³
<i>Holocarpa virgata</i>	273 ¹⁹⁰
<i>Hydropectis aquatica</i>	347 ²²⁶
<i>Hypochoeris radicata</i>	295, 324 ⁵²
<i>Inula japonica</i>	357, 358 ^{231,232}
<i>Inula viscosa</i>	310, ²⁰⁸ 308, 311, ²⁰⁵ 313, ²⁰⁹ 389, ²⁰⁹ 416, 342–344 ²²⁵
<i>Ixeris debilis</i>	368, 369, 373, 374, 380 ¹⁰⁸
<i>Jasonia candicans</i>	282 ³⁴
<i>Jasonia montana</i>	372, 349, ⁴⁸ 411 ²⁴⁴
<i>Laggara pterodonota</i>	338 ^{31,200}
<i>Laggara alata</i>	298, 299, 340, ⁸⁵ 277, 336, 341, ¹⁹⁴ 333, ²²² 337, ²²⁴
<i>Laggara pterodonta</i>	274, 312, 329, 353, ¹⁹¹ 286, ¹⁹¹ 287, ²⁰⁰ 288, 289, ¹¹ 332, ²²¹ 354, 355, ^{31,200} 412 ²⁴⁵
<i>Montanoa speciosa</i>	413 ²⁴⁸
<i>Onopordon acaulon</i>	384, 385, ²³⁸ 390 ²³⁹
<i>Onopordon illyricum</i>	391, 404 ²⁴⁰
<i>Perymenium featherstonei</i>	409, 410 ²⁴³
<i>Pluchea arguta</i>	418 ²⁵²
<i>Pluchea dioscoridis</i>	294, 403, ²⁰³ 400, 401 ⁴⁹
<i>Rudbeckia grandiflora</i>	321, 339 ²¹⁶
<i>Saussurea lappa</i>	360 ²³⁴
<i>Schistostephium rotundifolium</i>	309, 408 ²⁰⁶
<i>Sphaeranthus indicus</i>	320, ²¹⁴ 370 ¹³¹
<i>Tanacetum praeritum</i>	351, ²²⁸ 419 ²⁵³
<i>Taraxacum laevigatum</i>	371 ²³⁵
<i>Tessaria absinthioides</i>	356 ²³⁰
<i>Tessaria integrifolia</i>	316 ¹³⁸
<i>Ursinia eckloniana</i>	392 ³
<i>Ursinia tenuifolia</i>	328, 393 ³
<i>Verbesina turbacensis</i>	278 ¹⁹⁵
<i>Warionia saharae</i>	359 ²³³

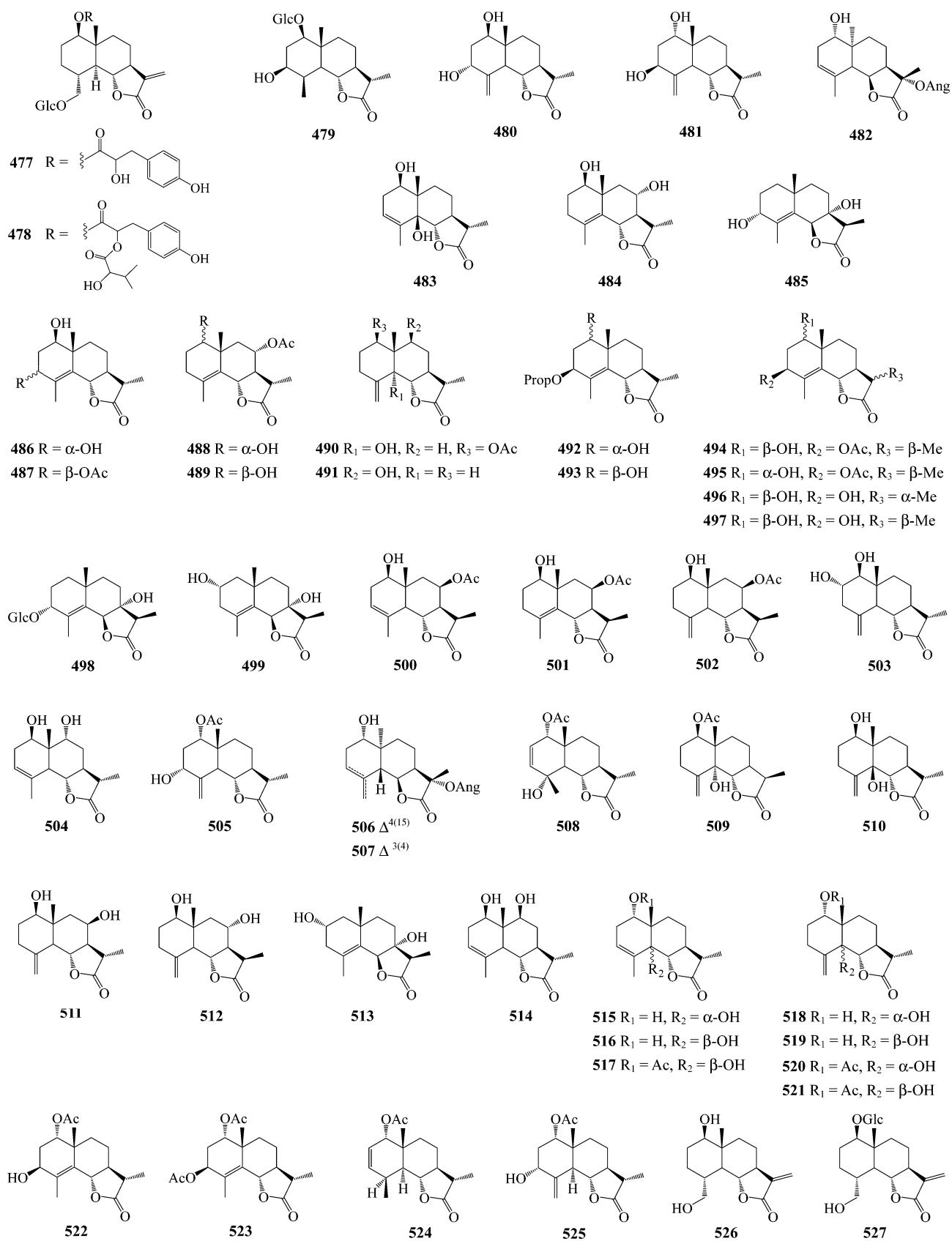
Spodoptera littoralis have been determined.⁴⁰³ A structure–activity relationship study of the sesquiterpene lactones that stimulate the germination of witchweed *Striga asiatica* has been described. The highest activity was observed with a mixture of santamarin and reynosin.⁴⁰⁴ The cytotoxic and antibacterial activities of the

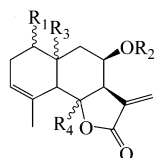
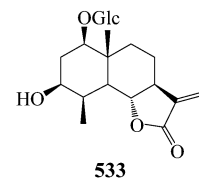
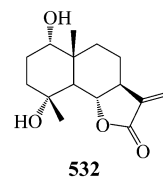
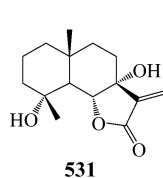
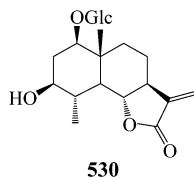
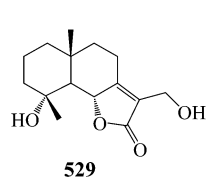
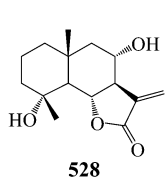
sesquiterpene lactones isolated from *Tanacetum praeritum* has been evaluated.⁴⁰⁵ The cytotoxic properties of the known eudesmane lactones from *Artemisia princeps* have been examined,⁴⁰⁶ and the pharmacological activity of reynosin^{407,408} and santamarine⁴⁰⁸ has been studied. The *in vitro* activity of compounds **687**, **688**, **689** and **695** against three Gram-positive and three Gram-negative bacteria was evaluated using a microdilution method, and their cytotoxic activity was determined against a panel of human tumour cell lines.³⁷⁴ Preliminary studies using the mycelial growth method indicated that the synthetic eudesmanolides **675**, **676**, **677**, **678** and **702** had inhibitory activities of against pathogenic fungi such as *Pythiumaphami dermatum*, *Phytophthora capsici*, *Fusarium oxysporum*, *Rhizoctonia solani* and *Botrytis cinerea* of 100–200 mg L⁻¹.⁴⁰⁹ Antiproliferative sesquiterpene lactones have been obtained from the roots of *Inula helenium*.⁴¹⁰ The effect of yomogin, an eudesmanolide with anti-inflammatory activity, has been investigated in human promyelocytic HL-60 leukaemia cells,⁴¹¹ whilst the antiplasmodial activity of the lactones isolated from *Eupatorium semialatum* has been studied.⁴¹² The antimicrobial activities of several lactones obtained from *Inula helenium*, *Rudbeckia subtomentosa*⁴¹³ and *Hemiteptia lyrata*⁴¹⁴ have been studied. The known lactones magnolialide and artemisin have been obtained from *Cichorium intybus*;⁴¹⁵ magnolialide has been shown to inhibit the growth of several tumour cell lines, but artemisin was inactive.⁴¹⁶ Studies of the inhibition of nitric oxide production by the lactone yomogin have been carried out.⁴¹⁷

The biotransformation of α -santonin by the fungus *Botrytis cinerea* afforded its 11 β -hydroxy derivatives in very good yield.⁴¹⁸ The microbiological transformation of deoxyvulgarin by *Rhizopus nigricans* and *Aspergillus ochraceus* has been reported,⁴¹⁹ and the microbiological transformation of α -santonin by *Pseudomonas cichorii*⁴²⁰ and *Aspergillus niger*⁴²¹ has also been studied. A biogenetic synthesis of vulgularin and peroxyvulgarin has been devised.⁴²² During the preparation of 4-*epi*-6 β -vulgarin, a biotransformation with the fungus *Rhizopus nigricans* was utilised in one of the synthetic steps.⁴²⁴ The biomimetic cyclisation of cnicin led to a new lactone, malactanolide **741**.⁴²⁵ The microbiological transformation of several sesquiterpene lactones by the fungi *Cunninghamella echinulata* and *Rhizopus oryzae* has been reported,⁴²⁶ and transformations of α -santonin, and α -, β -, and γ -cyclocostunolide with various fungi have been investigated.⁴²⁷ The microbiological transformations of 7 α -hydroxyfrullanolide⁴²⁸ and 11,13-dehydrosantonin⁴²⁹ by *Aspergillus niger* have been reported. The microbial transformations of 6 α - and 6 β -eudesmanolides by cultures of the fungus *Rhizopus nigricans* have been examined,⁴³⁰ and the microbiological transformation of α -santonin by several fungi has been studied.⁴³¹ A biomimetic synthesis of the bisessquiterpene lactones (\pm)-biatractylolide and (\pm)-biepiasterolide has been accomplished.⁴³² A *Pseudomonas* strain has been utilised in the biotransformation of α -santonin to 1,2-dihydrosantonin and the dihydroxy derivative **743**.⁴³³ The high photoreactivity of isoalantolactone with thymine to give photoadducts, involving the 5,6-double bond of thymine and the exomethylene double bond of the lactone, has been studied. This reaction may provide an explanation of the progressive evolution of contact dermatitis into chronic actinic dermatitis as an effect of the action of sesquiterpene lactones on the skin.⁴³⁴

A review of approaches towards the synthesis of *cis*-decalins has been reported. The *cis*-decalin framework is present in various classes of natural products.⁴³⁵ A review of the conversion of



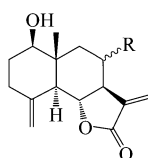




534 $R_1 = \alpha\text{-OH}$, $R_2 = \text{H}$, $R_3 = \alpha\text{-Me}$, $R_4 = \alpha\text{-H}$

535 $R_1 = \beta\text{-OH}$, $R_2 = \text{---} \text{C}(=\text{O})\text{CH}_2\text{CH}_2\text{OH}$,
 $R_3 = \beta\text{-Me}$, $R_4 = \beta\text{-H}$

536 $R_1 = \beta\text{-OH}$, $R_2 = \text{C}(\text{O})\text{Pr}^i$, $R_3 = \beta\text{-Me}$, $R_4 = \beta\text{-H}$



537 $R = \alpha\text{-OH}$

538 $R = \alpha\text{-OAc}$

539 $R = \alpha\text{-OAng}$

540 $R = \beta\text{-OH}$

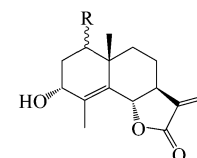
541 $R = \beta\text{-OC}(\text{O})\text{Pr}^i$

542 $R = \beta\text{-O} \text{---} \text{C}(=\text{O})\text{CH}_2\text{CH}_2\text{OH}$

543 $R = \beta\text{-O} \text{---} \text{C}(=\text{O}) \text{---} \text{C}(\text{CH}_3)_2 \text{---} \text{O}$

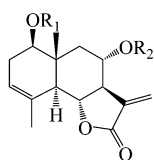
544 $R = \beta\text{-O} \text{---} \text{C}(=\text{O}) \text{---} \text{C}(\text{CH}_3)_2 \text{---} \text{C}(\text{OH})_2 \text{---} \text{CH}_3$

545 $R = \beta\text{-O} \text{---} \text{C}(=\text{O}) \text{---} \text{C}(\text{CH}_3)_2 \text{---} \text{C}(\text{OH}) \text{---} \text{CH}_3$



546 $R = \alpha\text{-OH}$

547 $R = \beta\text{-OH}$



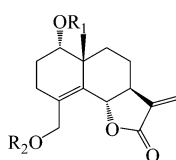
548 $R_1 = R_2 = \text{H}$

549 $R_1 = \text{Ang}$, $R_2 = \text{H}$

550 $R_1 = \text{H}$, $R_2 = \text{Meacr}$

551 $R_1 = \text{H}$, $R_2 = \text{C}(\text{O})\text{Pr}^i$

552 $R_1 = \text{H}$, $R_2 = \text{Ac}$

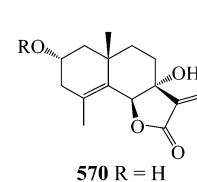
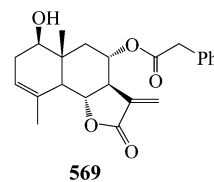
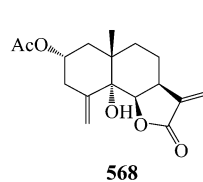
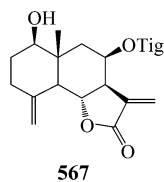
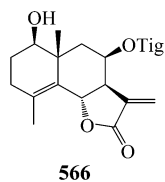
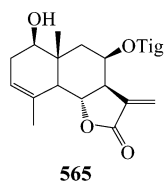
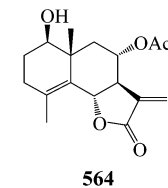
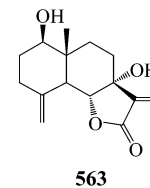
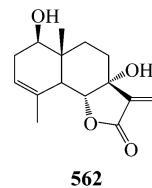
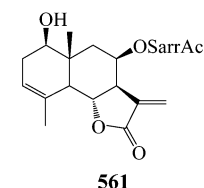
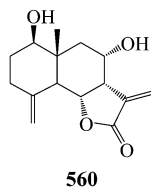
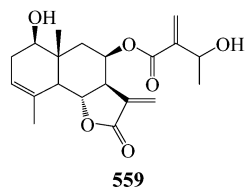
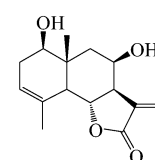
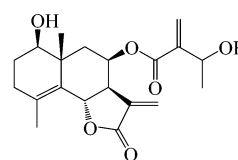


553 $R_1 = \text{H}$, $R_2 = \text{Meacr}$

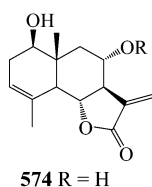
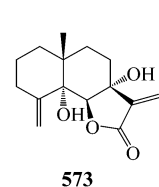
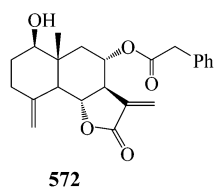
554 $R_1 = \text{H}$, $R_2 = \text{---} \text{C}(=\text{O})\text{CH}_2\text{CH}_2\text{OH}$

555 $R_1 = \text{Ac}$, $R_2 = \text{---} \text{C}(=\text{O})\text{CH}_2\text{CH}_2\text{OH}$

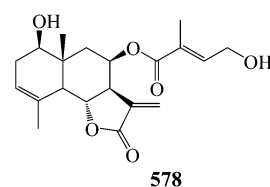
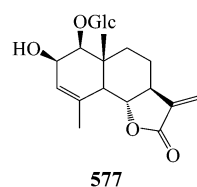
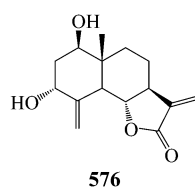
556 $R_1 = \text{H}$, $R_2 = \text{---} \text{C}(=\text{O}) \text{---} \text{C}(\text{CH}_3)_2 \text{---} \text{O}$

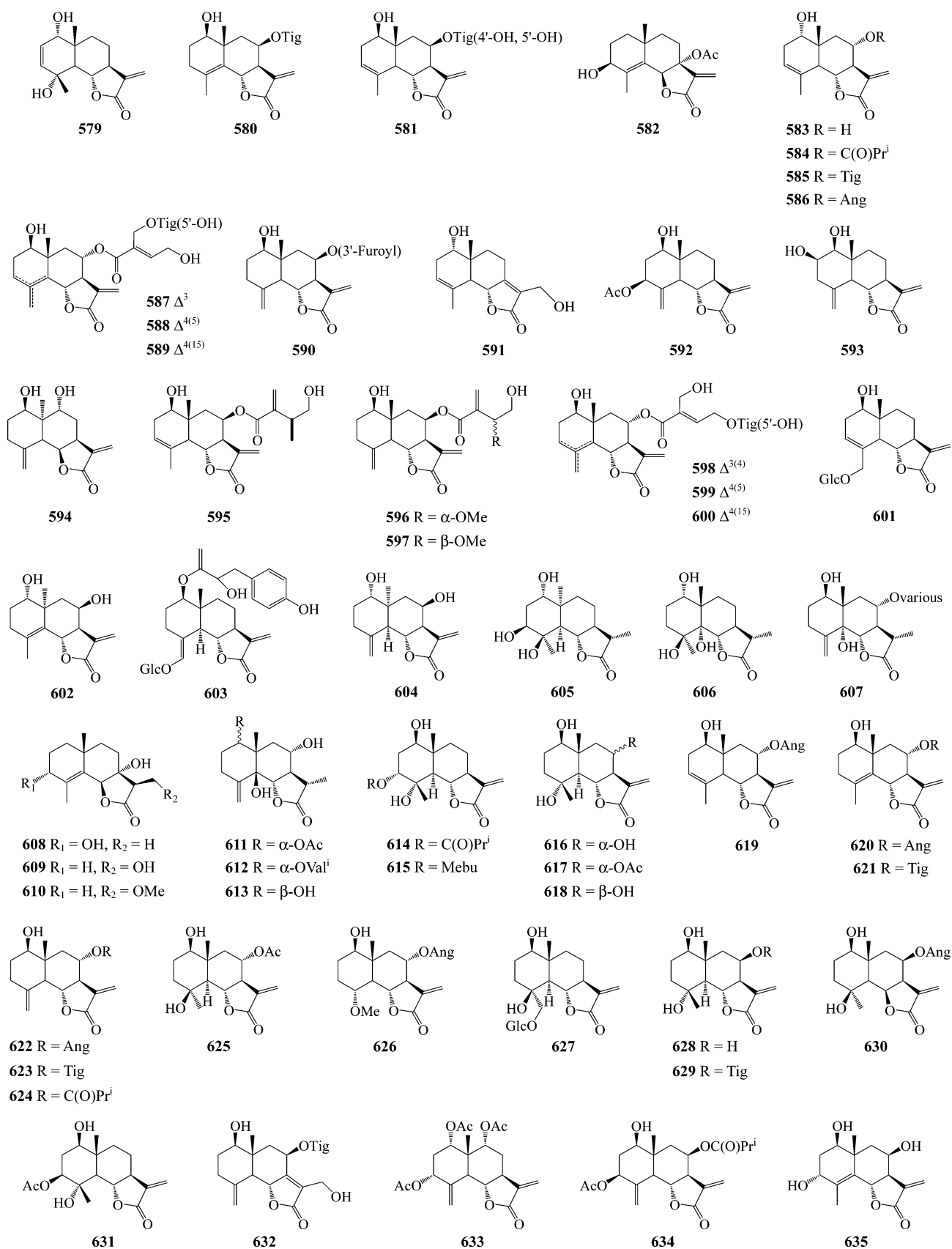


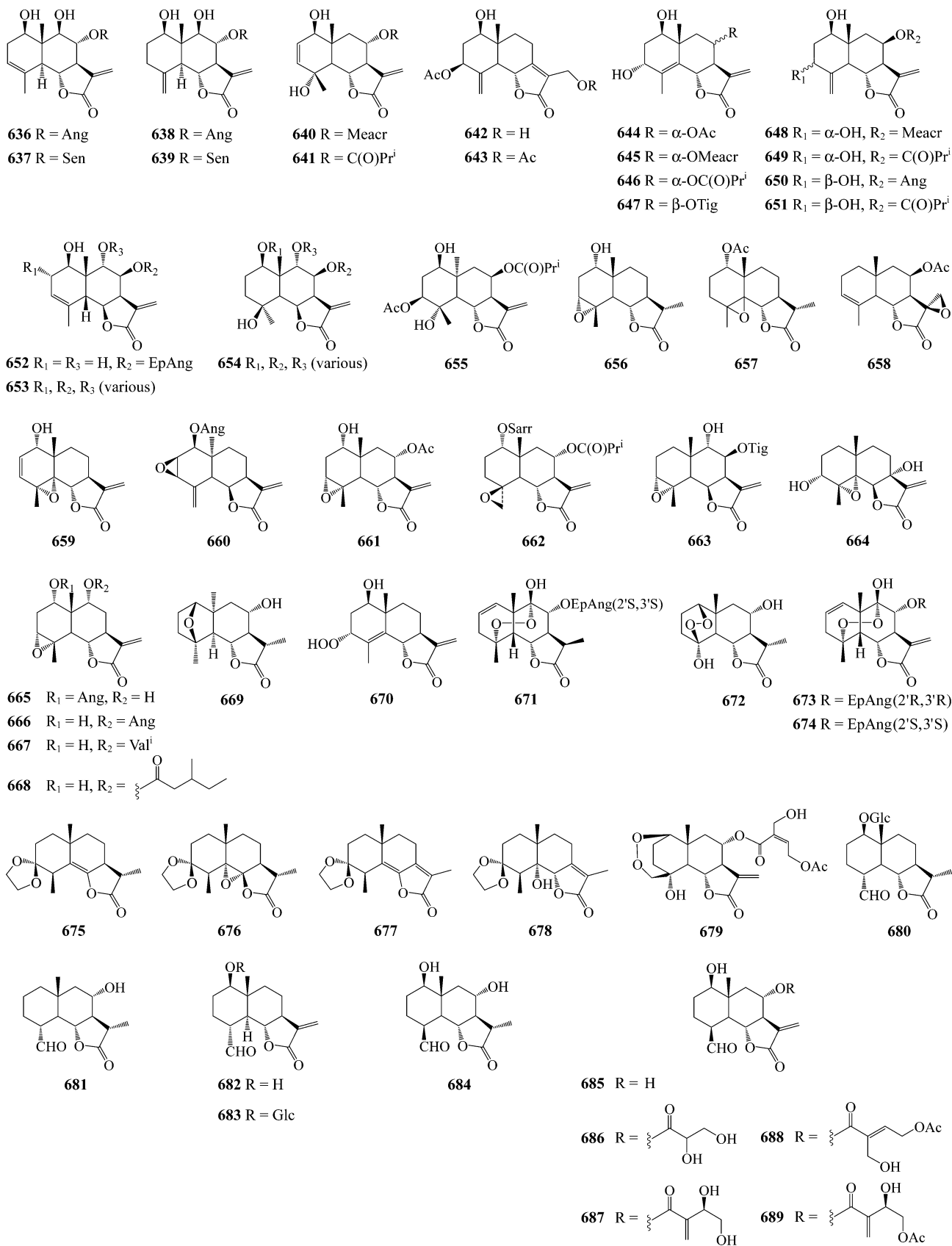
571 $R = \text{Ac}$

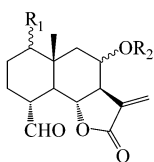


575 $R = \text{Ac}$



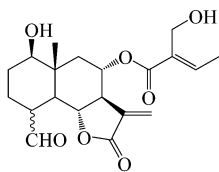




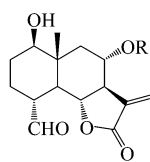


690 $R_1 = \alpha\text{-OH}$, $R_2 = \alpha\text{-Meacr}(4'\text{-OH})$

691 $R_1 = \beta\text{-OH}$, $R_2 = \beta\text{-Meacr}$

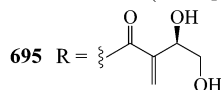


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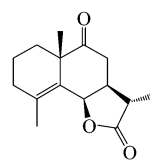


693 $R = \text{H}$

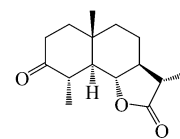
694 $R = \text{Meacr}(3'\text{-CH}_2\text{OAc}, 3'\text{-OH})$



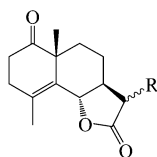
695 $R =$ [side chain]



696

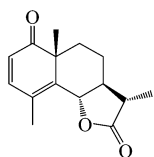


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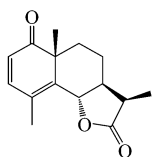


698 $R = \alpha\text{-Me}$

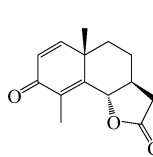
699 $R = \beta\text{-Me}$



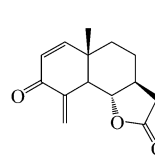
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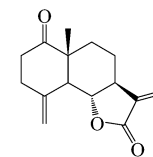
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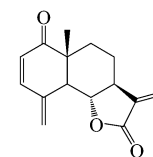
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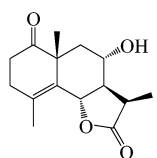
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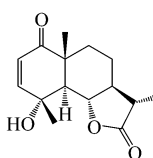
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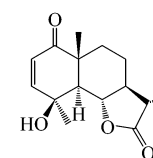
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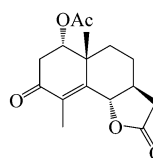
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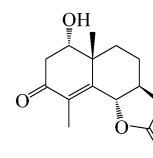
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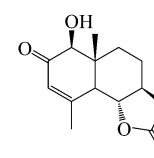
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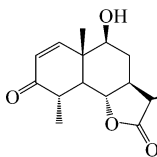
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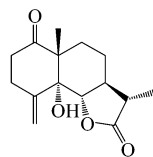
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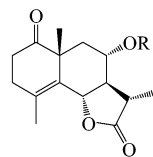
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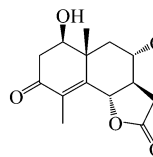
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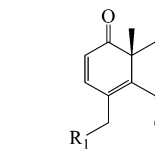
714 $R = \text{various}$



715 $R = \text{Ac}$

716 $R = \text{Meacr}$

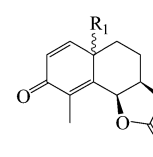
717 $R = \text{C(O)Pr}^i$



718 $R_1 = \text{OH}$, $R_2 = \text{H}$, $R_3 = \beta\text{-Me}$

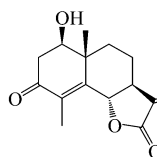
719 $R_1 = \text{OH}$, $R_2 = \text{H}$, $R_3 = \alpha\text{-Me}$

720 $R_1 = \text{H}$, $R_2 = \text{OH}$, $R_3 = \alpha\text{-Me}$

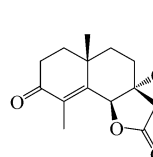


721 $R_1 = \alpha\text{-Me}$, $R_2 = \text{Ang}$

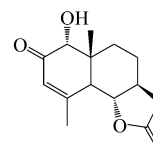
722 $R_1 = \beta\text{-Me}$, $R_2 = \text{H}$



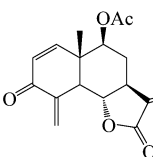
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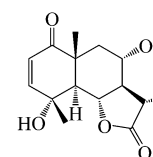
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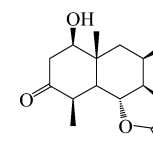
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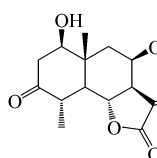
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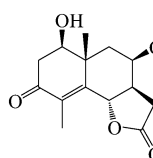
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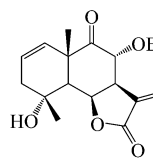
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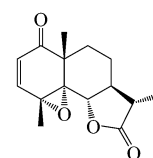
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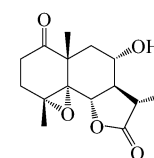
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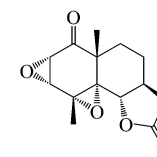
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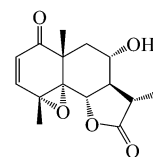
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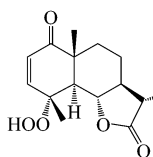
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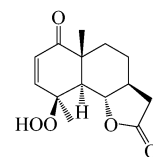
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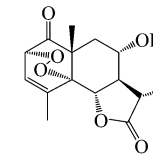
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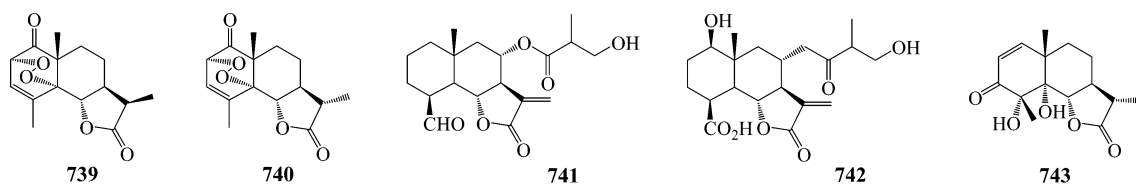
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α -santonin into other sesquiterpene lactones, such as eudesma-12,6-olides and eudesma-12,8-olides, has been published.⁴³⁶ Chemical, enzymatic and microbiological methods have been used in the transformation of santonin into sivasinolide and yomogin analogues.⁴³⁷ Ultrasound has been used to enhance the rate of the reductive cleavage of sesquiterpene γ -enone-lactones.⁴³⁸ An interesting procedure for the SmI₂-catalyzed rearrangement of a 5 α ,6 α -epoxy-11,13-dihydroeudesmanolide to an eudesm-5,7(11)-dienolide has been reported.⁴³⁹

The synthesis of deuterium-labelled eudesmanolides, starting from lactones isolated from *Inula helenium*, has been described.⁴⁴⁰ The partial syntheses of 7 β -hydroxyeudesmanolide,⁴⁴² erivanin, and 1-*epi*-erivanin⁴⁴³ has been reported. The syntheses of artepaulin, tuberin, oopodin, and vulgarin derivatives, starting from α -santonin, has been reported.⁴⁴⁴ The sesquiterpene lactone costunolide has been used in an efficient partial synthesis of several eudesmanolides.⁴⁴⁵ A synthesis of (+)-colartin and (+)-arbusculin together with their C-4 epimers has been devised, and the plant growth regulating activity of these lactones has been studied.⁴⁴⁶ A novel approach towards the stereocontrolled synthesis of eudesmanolides has been developed following a quasi-biomimetic strategy starting from a functionalised oxabicyclic template, by which the first total syntheses of gallicadiol and isogallicadiol were achieved.⁴⁴⁷ Another lactone, **722**, has been synthesised as a racemate,⁴⁴⁸ which implied a revision of the structure previously assigned.⁴⁴⁹ An enantioselective synthesis of (+)-decipienin A has been achieved.⁴⁵⁰ The syntheses, using free-radical chemistry, of (+)-3 α -hydroxyreynosin, (+)-reynosin and (+)-11 β ,13-dihydroreynosin, starting from 1,10-epoxy-11 β ,13-dihydrocostunolide, have been accomplished.⁴⁵¹

The syntheses of dehydrobrachylaenolide, isodehydrochamaecynone and *trans*-isodehydrochamaecynone, starting from tuberiferine, have been reported. Eudesmanolides possessing an α -methylene- γ -lactone moiety, such as dehydrobrachylaenolide, exhibited significant inhibitory activity toward the induction of the intercellular adhesion molecule-1 (ICAM-1).⁴⁵² The total synthesis⁴⁵³ of (+)-decipienin A **721** has been carried out, confirming the stereochemistry proposed by the Czech authors⁴⁵⁴ and correcting the original structure.⁴⁵⁵ (+)-Stoebenolide has been prepared starting from (+)-salonitenolide.⁴⁵⁶ A short synthesis of the sesquiterpene lactone 1-oxoeudesma-2,4-dien-11 β -hydro-12,6 α -olide has been achieved,⁴⁵⁷ and a synthesis of 11 β -angeloyloxy- α -santonin has been devised. In this work, the X-ray structure of 11 β -hydroxy- α -santonin was described.⁴⁵⁸ The partial synthesis of 6 β -eudesmanolides and 6 β -guaianolides from 6 α -eudesmanolides has been reported,⁴⁵⁹ and a synthesis of the reported structure of herbolide I has been devised, showing that the assigned structure is erroneous.⁴⁶⁰

A short synthesis of (+)-colarin and (+)-arbusculin, starting from α -santonin, has been accomplished.⁴⁶¹ The syntheses of the eudesmanolides torrentin and dihydrosantamarine, and the elemanolide saussaurea lactone have been reported,⁴⁶² and a synthetic

strategy whereby 6 β -eudesmanolide isomers are converted into the 6 α -analogues in modest yield has been developed.⁴⁶³ Artemisin has been used as the starting material in the preparation of herbolide E, showing that the previously proposed structure for this compound must be revised.⁴⁶⁴ Syntheses of 4 α ,5 α - and 4 β ,5 β -epoxy-eudesmanolides⁴⁶⁵ and the bioactive lactones (–)-arbusclin D **529** and (–)-*epi*-arbusclin D have been reported.³²³ A formal synthesis of (–)- α -santonin has been achieved using an optically active key intermediate, prepared by asymmetric hydrolysis of its racemate.⁴⁶⁶ Santonin and other sesquiterpene lactones react with pyrrolidine at room temperature to afford γ -hydroxyalkylamides,⁴⁶⁷ and the unexpected selective formation of a δ -lactone after the ozonolysis of a santonin derivative has been reported.⁴⁶⁸

A first total synthesis of (\pm)-diplophyllin has been devised,⁴⁶⁹ and a highly stereoselective synthesis of ivangulin has been achieved.⁴⁷⁰ A modified synthesis of racemic occidentaol has been reported,⁴⁷¹ as well as the synthesis and *in vitro* activity of 3-hydroxyencelin.⁴²⁵ An easy route to racemic 11-hydroxy-eudesmanolides, such as decipienin A, has been described,⁴⁷² and short syntheses of dehydroisoerivanin, isoerivanin, ludovicin C and 1 α ,3 α -dihydroxyarbusculin B have been described.⁴⁷³ (–)- α -Santonin has been used as starting material in the synthesis of phytuberin,⁴⁷⁴ and other studies towards the biomimetic synthesis of these sesquiterpene lactones have been described.⁴⁷⁵ Syntheses of (–)-isobaimuxinol and (–)-baimuxinol have been reported,^{476,477} and cytotoxic α -aminomethyl-substituted eudesmanolides have been prepared by stereoselective Michael-type addition of amines to alantolactone and isoalantolactone.⁴⁷⁸

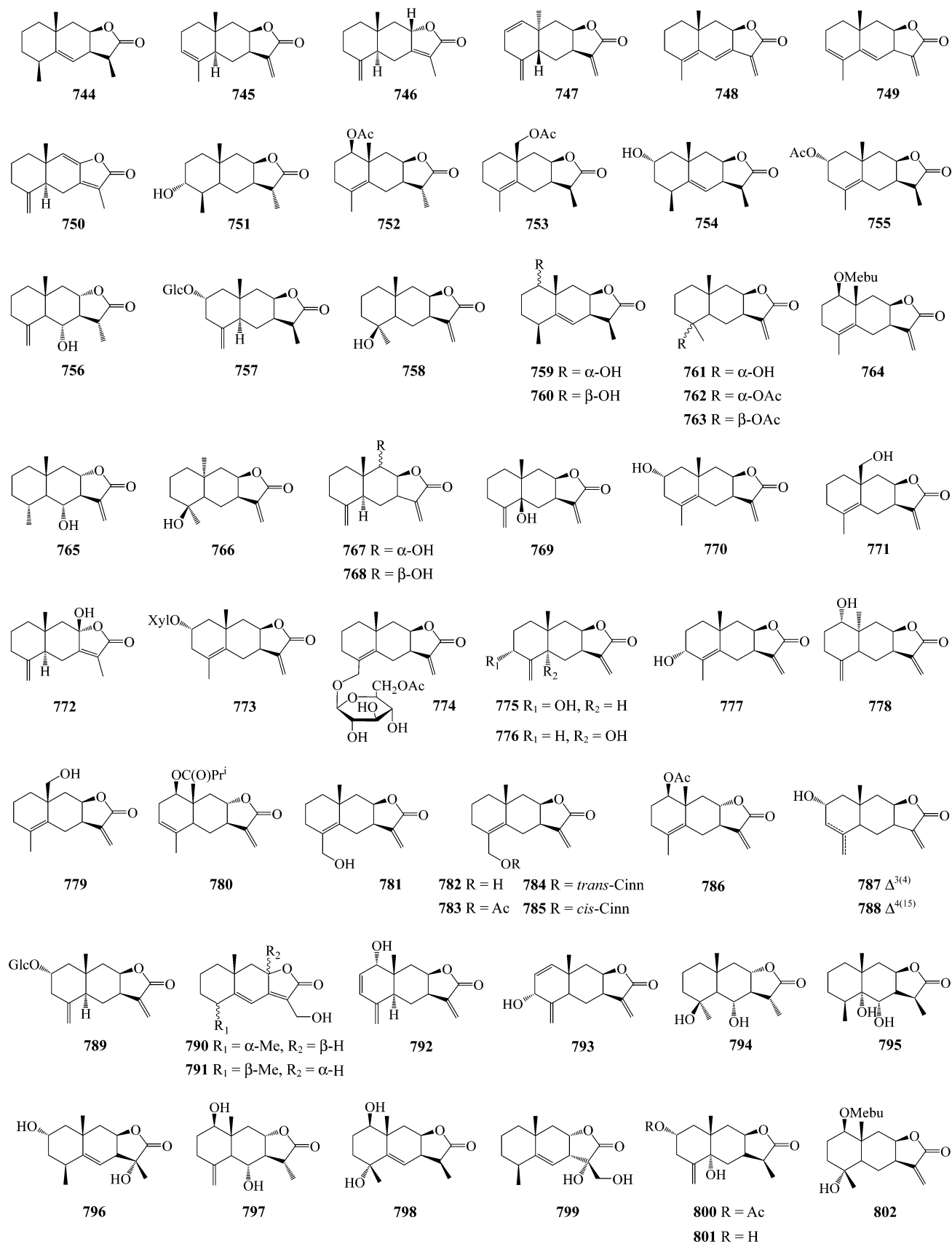
3.7.2 Eudesmane-12,8-olides. The new eudesmane-12,8-olides **744–917** and their plant sources are summarised in Table 9. Single crystal X-ray diffraction analysis has been used to establish the structures of sesquiterpene lactones **849**, **850** and **851**⁵²² obtained from *Steiractinia*, the telekin epimers **769** and **887**,⁵³⁸ as well as isogallicadiol,⁵⁴³ gazanniolide⁵⁴⁴ and compound **813**.⁵⁰⁸ The structure of eudesmanolide, isolated from *Lasiolanea santosi*⁵⁰³ and *Mikania goyazensis*,⁵⁰⁴ has been revised to **797**,⁵⁴⁵ and the stereochemistry of septuplinolide **761** has been revised, correcting the stereochemistry of the 4-OH group from β to α .⁴⁹⁰

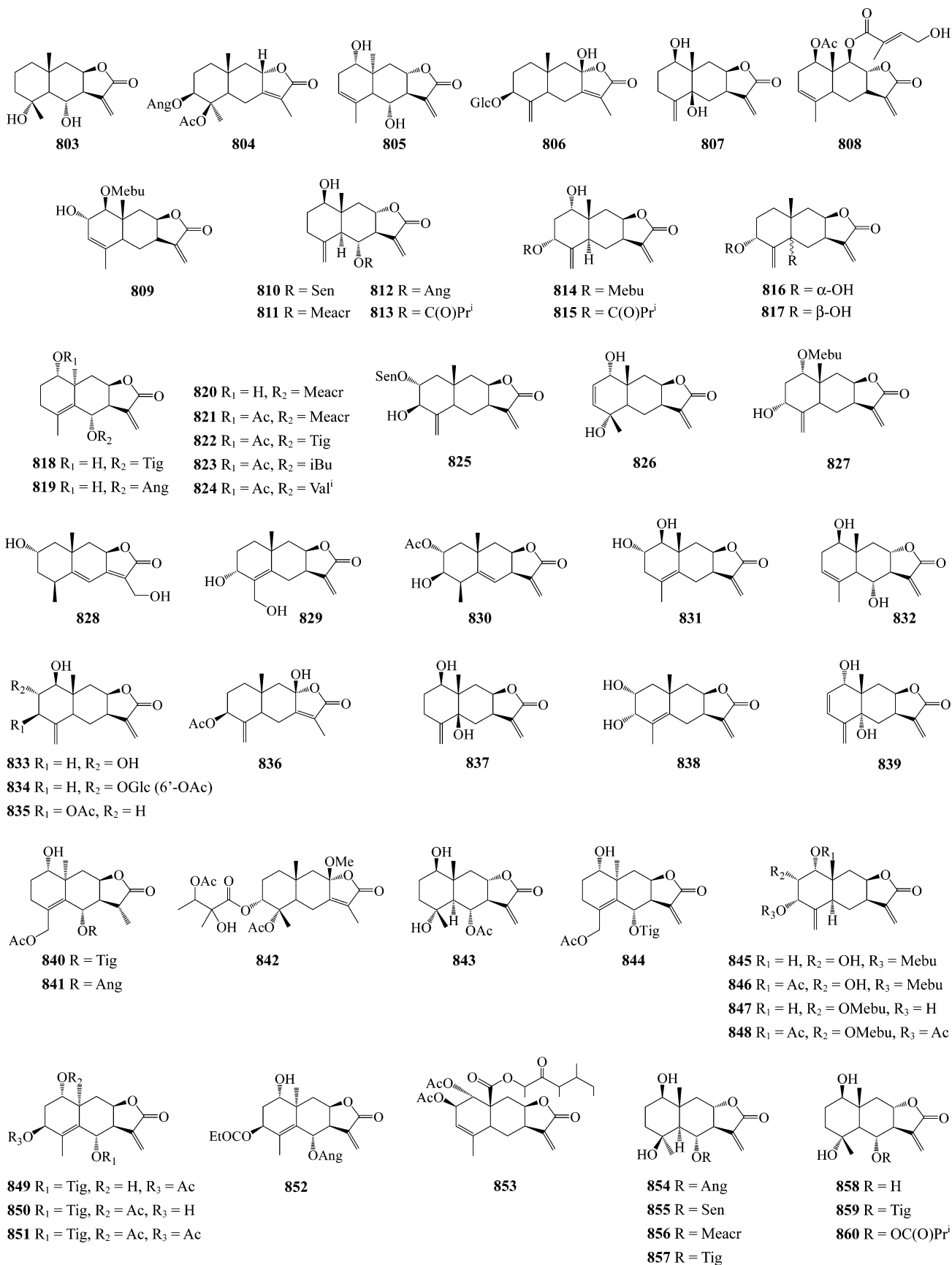
Inunal **745** and isoalantolactone **897** are biologically active as plant growth regulators, promoting the formation of adventitious roots.⁴⁸¹ The biotransformation of pyrenthrosin by *Rhizopus nigricans* has been investigated to generate eudesmanolide-type metabolites, which exhibited cytotoxic, antifungal, and antiprotzoal activities.⁵⁴⁶

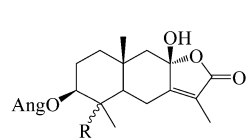
The chemical and microbiological transformation of (–)- α -santonin into 8-*epi*-artemisin has been carried out,⁴⁴¹ and the synthesis of 8-*epi*-ivangustin and 8-*epi*-isoivangustin starting from santonin has been explored.⁴⁴⁴ The synthesis of deuterium-labelled eudesmanolides, starting from lactones isolated from *Inula helenium*, has also been described.⁵⁴⁷ A partial synthesis

Table 8 Sources of eudesmane-12,6-olides

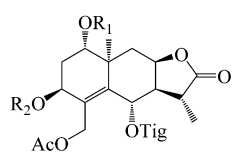
Sources	Compounds	Sources	Compounds
<i>Anthemis carpatica</i>	583–586 ³⁴⁵	<i>Grangea maderaspatana</i>	485 ³⁰⁸
<i>Arctotheca calendula</i>	703 ²⁴⁷	<i>Greenmaniella resinosa</i>	536, 541, 651, 728, 729 ³²⁸
<i>Argyranthemum foeniculaceum</i>	697 ³⁸⁰	<i>Gutenbergia cordifolia</i>	726 ²⁹⁴
<i>Artemisia afra</i>	643 ¹³⁶	<i>Helipterum roseum</i>	619–624, 626 ³⁵⁴
<i>Artemisia caerulescens</i>	488, 489, ¹³⁴ 669 ^{211,366}	<i>Hieracium intybaceum</i>	461, 467 ²⁹⁵
<i>Artemisia caerulescens</i> subsp. <i>gallica</i>	709 ³⁸⁷	<i>Inula graveolens</i>	460, 724 ²⁹³
<i>Artemisia caerulescens</i> var. <i>cretacea</i>	433 ²⁸⁰	<i>Inula salsoloides</i>	511 ³¹⁶
<i>Artemisia canariensis</i>	532, 575, 704, 705, ³²⁵ 727 ³⁹³	<i>Ixeris dentata</i>	465 ²⁹⁷
<i>Artemisia cretacea</i>	698 ³⁸¹	<i>Ixeris repens</i>	477, 478, 601, 603, 627 ¹⁰⁸
<i>Artemisia diffusa</i>	437, 710 ²⁸³	<i>Laser trilobum</i>	482, ³⁰⁶ 506, 507 ³¹⁴
<i>Artemisia fragrans</i>	438, ²⁸⁵ 475 ³⁰⁴	<i>Leucanthemopsis actinata</i>	429 ²⁷⁵
<i>Artemisia giraldui</i>	706 ³⁸⁵	<i>Liabum floribundum</i>	712 ³⁸⁹
<i>Artemisia gracilescens</i>	698 ³⁸²	<i>Liatris laevigata</i>	565–567, ³³⁹ 629 ²³⁹
<i>Artemisia herba-alba</i>	469, 481, 492, 493, ²¹⁸ 424, 426, 440, 470, 471, 699, ¹⁹⁸ 490, 491, ¹⁶¹ 700, 701, 718–720, 732, 734, 735, 738–740, ³⁸³ 427, 476, ²⁷² 429, 434, 436, 484, ²⁷⁶ 432, ²⁷⁹ 487, ³⁰⁹ 494–497, ³¹⁰ 514, ³¹⁹ 656 ³⁶¹	<i>Mikania guaco</i>	552, ³³⁶ 564, 644 ³³⁶
<i>Artemisia hugueti</i>	435, 437, 510, 515–523, 611 ²⁸²	<i>Montanoa frutescens</i>	636–639 ³⁵⁷
<i>Artemisia ifranensis</i>	606 ²⁸²	<i>Montanoa hibiscifolia</i>	671, 673, 674, 731 ³⁶⁷
<i>Artemisia inculta</i>	512 ³¹⁷	<i>Onopordon myriacanthum</i>	692 ³⁷⁶
<i>Artemisia judaica</i>	480, 486, 642, 707, ³⁰⁵ 708, 736, ³⁸⁶ 737 ³⁹⁴	<i>Onopordon ambiguum</i>	690 ³⁷⁵
<i>Artemisia lehmanniana</i>	524, 525 ³²¹	<i>Onopordon tauricum</i>	684, 742 ³⁷¹
<i>Artemisia lehmannianoda</i>	442 ²⁸⁸	<i>Pegolettia oxydonta</i>	534, 560 ⁸
<i>Artemisia lerchiana</i>	592, 631 ³⁴⁸	<i>Pegolettia senegalensis</i>	444–449, 534 ²⁸⁹
<i>Artemisia leucotricha</i>	514 ³²⁰	<i>Perymenium menzezii</i>	455 ²⁹⁰
<i>Artemisia lucentica</i>	425, 593 ²⁷³	<i>Picris aculeata</i>	462, 480, 486 ²⁹⁶
<i>Artemisia ludoviciana</i>	576 ³⁴²	<i>Picris spinifera</i>	711 ³⁸⁸
<i>Artemisia maritima</i>	483, ³⁰⁷ 547, 661, 715 ³³³	<i>Pluchea dioscoridis</i>	504, 548, 666–668, ³¹² 549, ³³⁴ 594, ⁴⁹ 665 ^{312,365}
<i>Artemisia nitrosa</i>	509 ³¹⁵	<i>Podochaenium eminens</i>	531 ³²⁴
<i>Artemisia pauciflora</i>	697 ³⁷⁹	<i>Pyrethrum santolinoides</i>	605 ³⁵⁰
<i>Artemisia pontica</i>	607, 612, 613 ³⁵¹	<i>Ratibida latipalearis</i>	659 ³⁶⁴
<i>Artemisia rutifolia</i>	439 ²²⁷	<i>Schistostephium crataegifolium</i>	538 ²⁰⁶
<i>Artemisia salina</i>	441 ⁸⁷	<i>Schistostephium heptalobum</i>	454, 546 ²⁰⁶
<i>Artemisia santolinifolia</i>	713 ³⁹⁰	<i>Schistostephium rotundifolium</i>	450 ²⁰⁶
<i>Artemisia santonicus</i>	714, 733 ³⁹¹	<i>Senecio chrysanthemoides</i>	505, 508 ³¹³
<i>Artemisia spicigera</i>	437 ²⁸⁴	<i>Sonchus asper</i>	472, 473, 527, 680, 683 ³⁰¹
<i>Artemisia splendens</i>	434 ²⁸¹	<i>Sonchus macrocarpus</i>	474, 526, ³⁰² 682 ³⁷⁰
<i>Artemisia tenuisecta</i>	657 ³⁶²	<i>Sonchus oleraceus</i>	430, 431, 451 ²⁷⁷
<i>Artemisia tournefortiana</i>	696 ²¹¹	<i>Sphaeranthus indicus</i>	428, 458, 462, ²⁷⁴ 457, ²⁹¹ 498, ¹⁶⁶ 513, ³¹⁸ 573, ³⁴¹ 582, 664, ¹³¹ 608–610 ³⁵²
<i>Athanasia calva</i>	569, 572 ³⁴⁰	<i>Sphaeranthus suaveolens</i>	459, 499, 568, 570, 571 ²⁹²
<i>Bartlettina karwinskiana</i>	658 ³⁶³	<i>Squamoappus skutchii</i>	539 ³²⁹
<i>Bishopanthus soliceps</i>	614, 615 ³⁵³	<i>Stevia aff. tomentosum</i>	500–502 ³¹¹
<i>Calea trichomata</i>	542, 545, 632, 650, ³³¹ 647 ³⁵⁹	<i>Stevia alpina</i>	580, 581 ³⁴⁴
<i>Calostephane divaricata</i>	630, 652–654, 663 ³⁵⁵	<i>Stevia breviaristata</i>	578 ³⁴³
<i>Cassinia subtropica</i>	558, 635 ¹⁶	<i>Tanacetum ferulaceum</i>	528 ³²²
<i>Cassinia uncata</i>	463, 464, 537, 538, 540, 574, 602, 604, 616–618, 625, 730 ¹⁶	<i>Tanacetum parthenium</i>	540, ³³⁰ 579, 591, 670, ²²⁸ 725 ²⁵³
<i>Centaurea aspera</i>	672, 679 ³⁶⁸	<i>Tanacetum vulgare</i>	723 ³⁹²
<i>Centaurea attica</i>	687 ²⁴¹	<i>Taraxacum hallaisanensis</i>	479, 530 ³⁰³
<i>Centaurea deusta</i>	694 ³⁷⁸	<i>Taraxacum</i>	533, 577 ³²⁶
<i>Centaurea granata</i>	681 ³⁶⁹	<i>linearisquameum</i>	
<i>Centaurea malacitana</i>	686, 741 ³⁷³	<i>Tithonia diversifolia</i>	634, ³⁵⁶ 655 ³⁶⁰
<i>Centaurea ornata</i>	432 ²⁷⁸	<i>Tithonia rotundifolia</i>	543, 544, 557, 559, 561 ³³²
<i>Centaurea spinosa</i>	687, 688, 689, 695 ³⁷⁴	<i>Ursinia abrotanoides</i>	628 ³
<i>Centaurea thessala</i>	685 ^{241,372}	<i>Vladimiria souliei</i>	469, 503 ³⁰⁰
<i>Centaurea tweediei</i>	691 ²⁵⁷	<i>Zexmenia lantanifolia</i>	535 ³²⁷
<i>Centaurea zuccariniana</i>	693 ³⁷⁷		
<i>Chamaemelum fuscum</i>	550, 551, 645, 646, 716, 717, ³³⁵ 640, 641, 648, 649 ³⁵⁸		
<i>Cratystylis conocephala</i>	633 ²¹⁵		
<i>Decachaeta ovatifolia</i>	562, 563 ³³⁸		
<i>Dendroseris neriifolia</i>	468 ²⁹⁹		
<i>Dicoma macrocephala</i>	466 ²⁹⁸		
<i>Dimerostemma asperatum</i>	662 ³³⁷		
<i>Dimerostemma bishoppii</i>	553–556 ³³⁷		
<i>Disynaphia multicrenulata</i>	590 ³⁴⁷		
<i>Echinops spinosissimus</i>	440 ²⁸⁶		
<i>Eupatorium semialatum</i>	587–589, 598–600, ³⁴⁶ 595–597 ³⁴⁹		
<i>Geigeria rigida</i>	452, 453, 456, 534, 560 ²¹⁷		



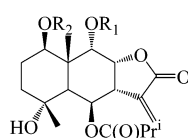




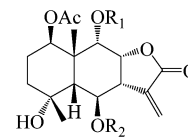
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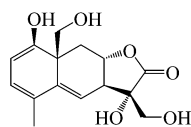
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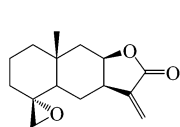
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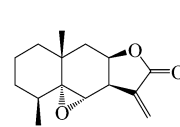
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868 R₁ = H, R₂ = Meacr
869 R₁ = Ac, R₂ = C(O)Pr^d
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871 R₁ = C(O)Pr^d, R₂ = Ac
872 R₁ = Ac, R₂ = C(O)Pr^d



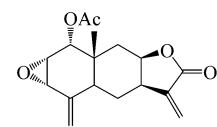
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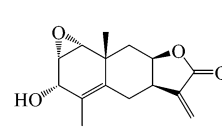
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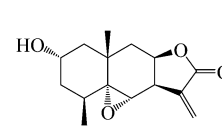
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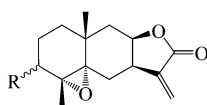
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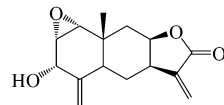
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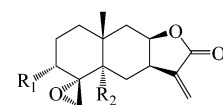
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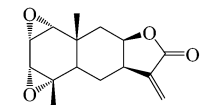
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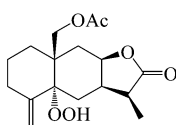
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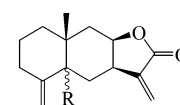
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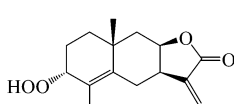
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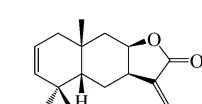
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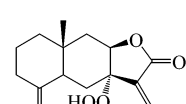
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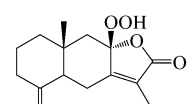
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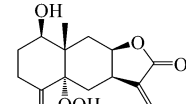
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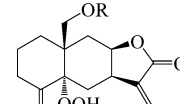
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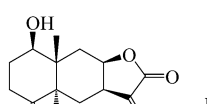
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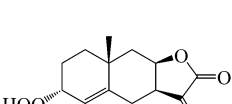
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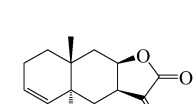
893 R = H
894 R = Ac



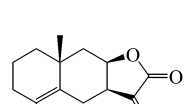
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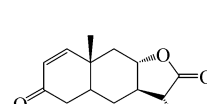
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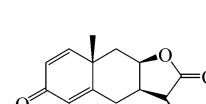
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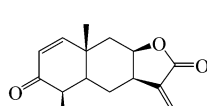
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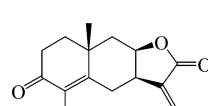
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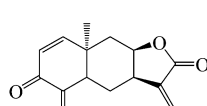
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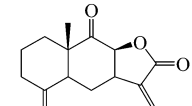
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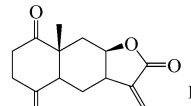
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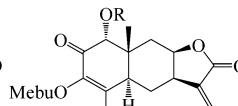
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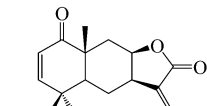
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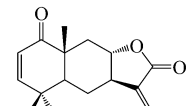
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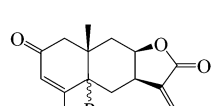
906 R = H
907 R = Ac



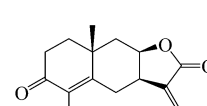
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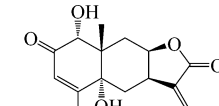
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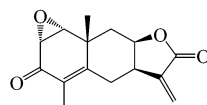
910 R = α -OH
911 R = β -OH



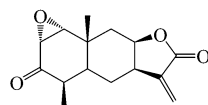
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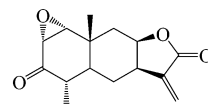
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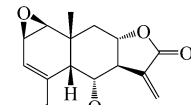
914



915



916



917

Table 9 Sources of eudesmane-12,8-olides

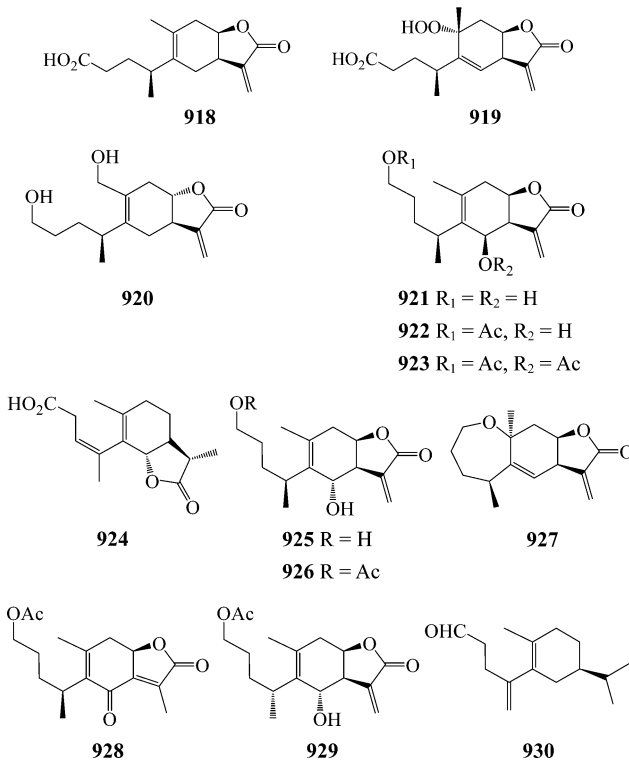
Sources	Compounds
<i>Ambrosia artemisioides</i>	782, 783, 784, 785, 829, 874, 879, 896, 912 ³⁸
<i>Anthemis altissima</i>	805 ⁵⁰⁶
<i>Artemisia argyi</i>	762, 763 ⁴⁹¹
<i>Artemisia feddei</i>	884, 536, 915 ⁵⁴²
<i>Artemisia iwawayomogi</i>	745, 775–777, 816, 817, 879, 880, 888, 889, 902 ²⁸⁰
<i>Artemisia umbelliformis</i>	886, 887 ⁵³⁷
<i>Artemisia xerophytica</i>	826 ⁵¹²
<i>Aspilia plurisetia</i>	849–851 ⁵²²
<i>Atractylodes japonica</i>	746, 750, 772, 107, 806 ⁵⁰⁷
<i>Atractylodes macrocephala</i>	836, 519, 891 ⁵³⁹
<i>Bahia absinthifolia</i>	774 ⁴⁹⁷
<i>Blumea afrakiana</i>	827 ⁵¹³
<i>Blumea densiflora</i>	814, 815, 845–848, 906, 907 ⁵⁰⁹
<i>Calea septuplinerva</i>	761, 490, 758 ⁴⁸⁸
<i>Calea subcordata</i>	780, 498, 813, 508, 909 ⁵⁴¹
<i>Carpesium cernuum</i>	791 ^{501,502}
<i>Carpesium macrocephalum</i>	755, 800, 801, 485, 757, 789, 487, 755, 795 ⁴⁸⁶
<i>Cassinia subtropica</i>	843 ¹⁶
<i>Eriocephalus africanus</i>	837, 885, 892–895 ²⁰²
<i>Eriocephalus pauperrimus</i>	771, 807 ²⁰²
<i>Eriocephalus scariosus</i>	752–753, 779 ²⁰²
<i>Ferreyranthus fruticosus</i>	792, 793, 839, 877, 881, 884, 900, 908, 913, 914, 916 ²⁵¹
<i>Flourensia macrophylla</i>	767, 768, 904, 905 ²⁰⁴
<i>Flourensia riparia</i>	766 ⁴⁹⁴
<i>Gnephosis arachnoidea</i>	786, 831 ⁵⁰⁰
<i>Gochmatia vernonioides</i>	857 ⁵²⁵
<i>Greenmaniella resinosa</i>	910, 911 ³²⁸
<i>Guizotia scabra</i>	876 ⁵³³
<i>Hyaloseris salicifolia</i>	833, 834 ⁵¹⁷
<i>Inula britannica</i>	803 ⁵⁰⁵
<i>Inula caspica</i>	825 ⁵¹¹
<i>Inula japonica</i>	760 ²³¹
<i>Inula macrophylla</i>	744, 873, 479, 744, 480, 790, 799 ⁴⁷⁹
<i>Inula montana</i>	835 ⁵¹⁸
<i>Inula racemosa</i>	745, 481, 748, 749, 483, 781, 874, 875, 898, 499, 882, 883, 890, 535, 897 ⁴⁸¹
<i>Inula thapsoides</i>	759, 760, 798 ⁴⁸⁹
<i>Lasiolanea santosii</i>	797 ⁵⁰³
<i>Mikania cynanchifolia</i>	917 ⁵⁵¹
<i>Mikania goyazensis</i>	797 ⁵⁰⁴
<i>Ondetia linearis</i>	754, 787, 788, 796 ¹⁰⁹
<i>Pegolettia oxydonta</i>	808 ⁸
<i>Perymenium featherstonei</i>	901 ²⁴³
<i>Pluchea dioscordis</i>	773 ⁴⁹⁶
<i>Pluchea quitoc</i>	804, 861, 862 ¹⁰¹
<i>Pulicaria crispa</i>	878 ⁵³⁴
<i>Pulicaria undulata</i>	770, 828 ⁴⁹⁵
<i>Schistostephium crataegifolium</i>	810–813, 854–856 ²⁰⁶
<i>Senecio flammeus</i>	853 ⁵²⁴
<i>Spilanthes acmella</i>	899 ⁵⁴⁰
<i>Spilanthes leiocarpa</i>	747 ⁴⁸²
<i>Stevia achalensis</i>	751, 778, 903 ⁴⁸⁴
<i>Tanacetopsis mucronata</i>	832 ⁵¹⁵
<i>Tanacetum argenteum</i>	859 ⁵²⁷
<i>Tanacetum densum</i>	832, 516, 837, 520, 858 ⁵²⁶
<i>Tanacetum ferulaceum</i>	756, 794, 322, 765 ⁴⁹³
<i>Tanacetum parthenium</i>	859 ⁵²⁹
<i>Tessaria ambigua</i>	842 ¹³⁸
<i>Vicoa pentanema</i>	830, 838 ⁵¹⁴
<i>Wedelia grandiflora</i>	818–824 ⁴¹⁰
<i>Wedelia hispida</i>	863, 864 ⁵²⁸
<i>Wedelia paludosa</i>	871, 872 ⁵³²
<i>Wedelia pinetorum</i>	840, 841, 844 ⁵²¹
<i>Wedelia prostata</i>	867–870 ⁵³¹
<i>Wedelia prostrata</i>	865, 866 ⁵³⁰
<i>Wunderlichia mirabilis</i>	764, 802, 809 ⁴⁹²
<i>Zexmenia virgulata</i>	852 ⁵²³

of (+)-ivalin has been achieved,⁴²³ and a general procedure for the synthesis of both 12,6- and 12,8-eudesmanolides has been described.⁵⁴⁸ Partial syntheses of (+)-isoalantolactone and (+)-isoalloyalantolactone have been devised,⁵⁴⁹ and the synthesis of (–)-dehydrobaimuxinol has been reported.⁵⁵⁰

4 Secoeudesmanes

4.1 1,10-Secoeudesmanes

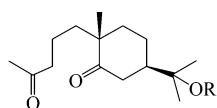
1,10-Secoeudesmanes **918** and **919** have been found in *Gnephosis arachnoidea*.⁵⁰⁰ Metabolite **920** has been isolated from *Calostephane divaricata*,⁵⁵⁵ **921** and **923** have been identified from *Inula britannica*,⁵⁵² and the secoeudesmanes **922**, **925** and **926** have been found in *Inula japonica*.⁵⁵³ Compound **924** has been isolated from *Artemisia hugueti*,²⁸² **927–929** have been found in *Inula britannica*,⁵⁵⁴ and **930** has been isolated from *Solidago gigantea*.¹²⁹



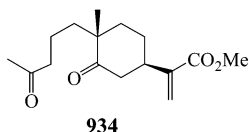
Studies on the semi-synthesis, structure determination and biotransformation of cyclic sulfinated eudesmane derivatives have been described.⁵⁵⁵ The structures of deacetylinulicin and inulicin, two secopseudoguaianolides extracted from *Inula japonica*, have been revised to the secoeudesmanolides **921** and **922** respectively.^{552,553} An enantioselective synthesis of the 1,10-*seco*-eudesmanolides eriolanin and eriolangin has been achieved.⁵⁵⁶

4.2 4,5-Secoeudesmanes

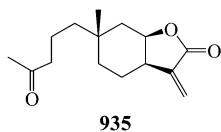
4,5-Secoeudesmanolide **935**⁴⁸⁰ has been identified from *Inula macrophylla*. The diketone eudesmanes **931**,²⁰² **932**,⁷ **933**¹⁰⁸ and **934**³ have been found in *Eriocephalus africanus*, *Iphiona scabra*, *Ixeris repens* and *Ursinia eckloniana*, respectively. 6,7-Secoeudesmane, (–)- α -eudesmol⁵⁵⁷ and 7-*epi*- γ -eudesmol⁵⁵⁸ have been described, and a racemic synthesis of balanitol has been reported.⁶⁹ The enantioselective total syntheses of chrysanthemol,^{559,560}



- 931 R = H
 932 R = Xyl
 933 R = Xyl(2'-OAc)



934

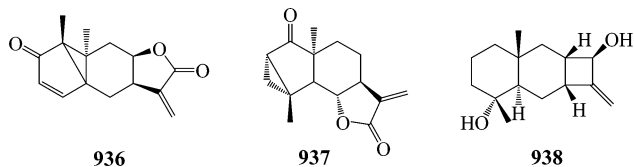


935

4,5-dioxo-*seco*- γ -eudesmol, 5 β ,11-dihydroxyiphionan-4-one⁵⁶¹ and the secoeudesmane glutinone,^{320,562} have been reported. The total syntheses of 4,5-secoeudesmane-type sesquiterpenes, such as 4,5-dioxo-10-*epi*-4,5-*seco*- γ -eudesmane and 4,5-dioxo-10-*epi*-4,5-*seco*- γ -eudesmanol, have also been reported.⁵⁶³

5 Cycloeudesmanes and isomeric eudesmanes

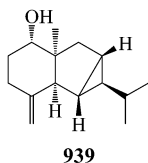
The cycloeudesmanes are few in the Asteraceae family. For example, the methyl-transferred 1,5-cycloeudesmane **936** has been identified from *Ferreyranthus fruticosus*²⁵¹ and 2,4-cycloeudesm-12,6-olide **937**, the structure of which was confirmed by X-ray analysis, is a constituent of *Ratibida latipalearis*.^{564,565} 8,12-Cycloeudesmene **938** has been isolated from *Jasonia candicans*,³⁴ 6,8-cycloeudesmene **939** has been identified from *Solidago gigantea*,¹²⁹ and 2,4-cycloeudesm-12,8-olide **940** is a constituent of *Ondetia linearis*.¹⁰⁹ The isomeric eudesma-13-acids **941**⁵⁶⁶ and **942**³⁶⁰ have been found in *Tithonia diversifolia*, and **943–947** were obtained from *Inula macrophylla*.⁴⁸⁰



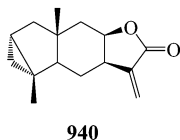
936

937

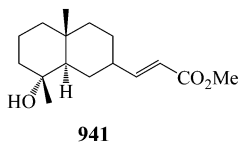
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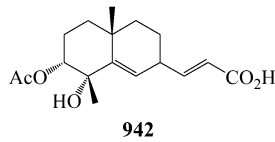
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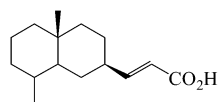
940



941



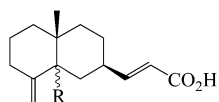
942



943 $\Delta^{4(5)}$

944 4 α ,5 α -epoxy

945 4 β ,5 β -epoxy



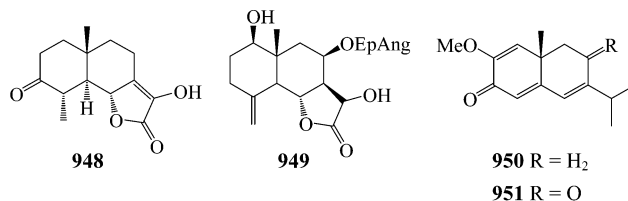
946 R = α -OH

947 R = β -OH

6 Noreudesmanes

15-Noreudesmanes **950** and **951** are elaborated by *Parthenium argentatum*,¹³⁷ and eudesmanes **952** and **958** have been isolated from *Apalochlamys spectabilis*¹⁹⁷ and *Microlabium polymniodes*³⁸⁴ respectively. Sesquiterpene **959** has been elucidated from *Onopordon ambigum*,²⁵⁶ and compound **957** is a constituent of *Erigeron annuus*.³⁹ A stereoselective synthesis of the norsesquiterpene platyphyllide has been achieved.⁵⁶⁷

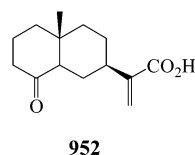
14-Noreudesmanes **953**, **954**⁵⁶⁸ and **956**¹³³ have been identified from *Ligularia dentata*, and **955** has been isolated from *Senecio gilliesianus*.⁵⁶⁹ The 13-noreudesmanes **948** and **949** have been found in *Crepis pygmaea*⁵⁷⁰ and *Tithonia pedunculata*,⁵⁷¹ respectively. The norsesquiterpenolide **948** has been synthesised from α -santonin.⁵⁷² Di-noreudesmane **960** has been isolated from *Jasonia montana*,⁴⁸ and trinoreudesmanes **961** and **962** have been found in *Pluchea arguta*²⁵² and *Rebutia marsoneri*,⁵⁷³ respectively. A practical synthesis of enantiomerically pure (–)-geosmin has been achieved,⁵⁷⁴ and the eudesmane sesquiterpene (+)-dehydrogeosmin **962** has been prepared by synthesis.⁵⁷⁵ The biosynthesis of the trinorsesquiterpene geosmin in a *Streptomyces* species and also in the liverwort *Fossombronina pusilla* has been investigated. This work revealed that the bacterium produces geosmin *via* 1-deoxy-D-xylose, whilst the liverwort uses the mevalolactone pathway.⁵⁷⁶ An efficient procedure for creating precise gene replacements in *Streptomyces coelicolor* has permitted the identification of a protein domain required for the biosynthesis of the trinorsesquiterpene geosmin,



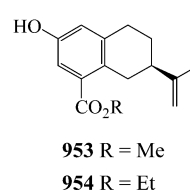
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949

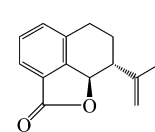
950 R = H₂
 951 R = O



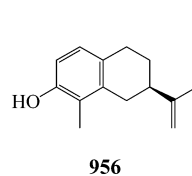
952



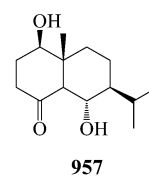
953 R = Me
 954 R = Et



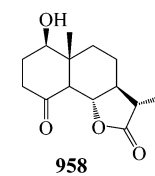
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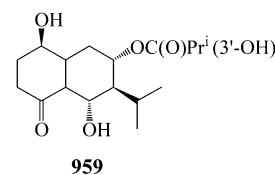
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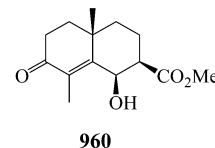
957



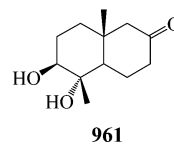
958



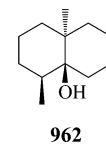
959



960



961



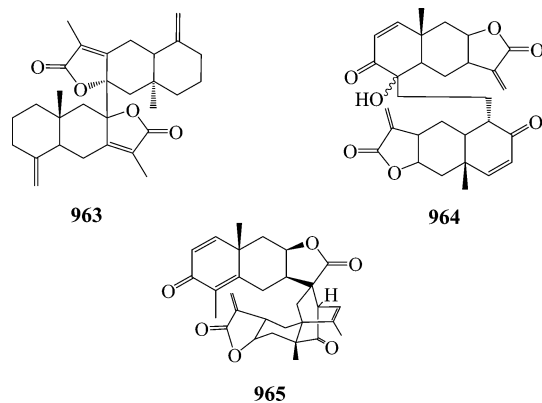
962

the metabolite responsible for the musty-earthy odour of soil.⁵⁷⁷ Moreover, the expression and mechanistic analysis of a germacadienol synthase from this *Streptomyces* species, implicated in the biosynthesis of geosmin, has been described.⁵⁷⁸ The quantitative determination of this norsesquiterpene in red beets and wheat grain⁵⁷⁹ has also been reported.

7 Dimers

7.1 Dieudesmanes

The bis-sesquiterpene biatractylode **963** has been found in an extract of the Chinese medicinal herb *Atractylodes macrocephala*.^{580,581} A biomimetic synthesis of the bis-sesquiterpenes biatractylolide and biepiasterolide has been achieved.^{582,583} A dimeric eudesmanolide, hydroxyl-bis-dihydroencelin **964**, has been found as a component of the aerial parts of *Montanoa speciosa*,⁵⁸⁴ and the structure of 2(11'),5(13')-dieudesmane **965** from *Ferreyranthus fruticosus* has been elucidated.²⁵¹ The structure of the dimeric lactone biatractylolide has been determined by X-ray analysis.⁵⁸⁵



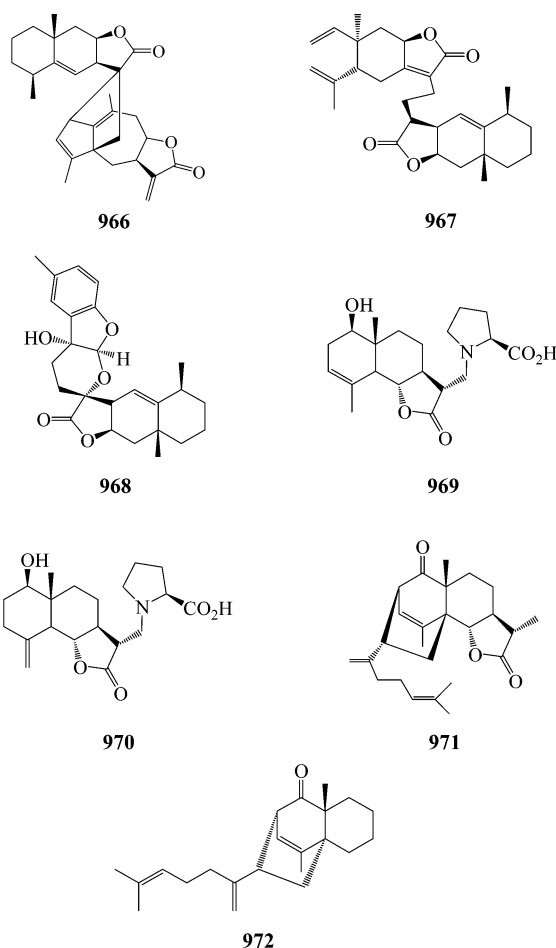
7.2 Iso-dimers

Rudbeckiolide **966**, a new dimeric sesquiterpene lactone with a eudesmane and a guaiane skeleton, has been obtained in low yield from *Rudbeckia laciniata*.⁵⁸⁶ Compound **968** and macrophyllidimer A **967** are two novel sesquiterpenes, which have been found in an extract from the bark of *Inula macrophylla*.^{587,588} Sausureamines D (**969**) and E (**970**), formed from a eudesmanolide and an alkaloid, have been isolated from *Saussurea lappa*.⁵⁸⁹ Two new compounds, **971** and **972**, adducts of a eudesmene lactone with a monoterpene, and of a trinoreudesmane with a monoterpene, have been obtained from *Artemisia herba-alba*.³⁸³

8 Conclusions

The Asteraceae family includes many species with a variety of uses, some of which are economically important.¹ The majority of eudesmanolides described in this report were isolated from either the aerial parts or the whole plants, mainly by "traditional" methods, but also by HPLC. The stereochemistry of many compounds was determined conclusively – molecular mechanics methods were also used in a number of cases to calculate conformations and relative stabilities.⁵⁹⁰

The 972 compounds described in this review were isolated from 247 species (of 110 genera), 523 of these compounds belonging to



the eudesmanolide group – a field in which Chinese researchers have been very active. The phytochemistry studies were focused on the large genera: *Artemisia*, *Centaurea*, *Inula*, *Pluchea*, etc. Our literature search has shown that interest in eudesmanoids from the Asteraceae is decreasing (see Fig. 1), although many researchers are still engaged in this area. For the period 1983–1989, the number of eudesmanes reported from the Asteraceae species was 75% of the number from the other higher plants and marine organisms.

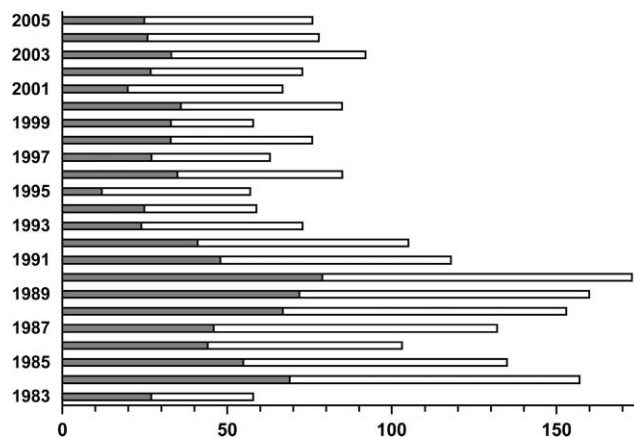


Fig. 1 Distribution of the total number of eudesmanes from 1983 to 2005. The grey part of each bar shows the number of eudesmanes reported from the Asteraceae species. The white part of each bar shows the number of eudesmanes reported from the other higher plants and marine organisms.

For the period 1990–1999, this proportion fell to 65%, and for the period 2000–2005, it fell again to 50%.

In the early years of eudesmanoid research there was less emphasis on biological testing, but increasingly there has been a focus on the biological properties of these compounds. Therefore the starting point for many investigations is the use of plants in traditional medicine, followed by the isolation and biological testing of their active principles. The broad and often very promising biological properties of eudesmanoids are very well known. Studies on the semi-synthesis, total synthesis and biotransformation of eudesmane derivatives have been described and have been increasing in number. Structure–activity relationships of sesquiterpene lactones have also been undertaken.⁵⁹¹ However, systematic structure–activity studies with cell cultures or other biological models, which might give insight into the mechanism of activity of eudesmanoids on the molecular level, seem to be absent. This might be a promising direction in which work in the field of eudesmanoids from the Asteraceae may proceed.

Recently, there has been great interest in the study of chemotaxonomy based on secondary metabolites from the Asteraceae family. In these studies, sesquiterpene lactones, which are used as taxonomic markers, are the most studied class of secondary metabolites. More than 4000 sesquiterpene lactones with around 30 different skeletons have so far been reported from several tribes of Asteraceae,⁵⁹² from which it is clear that eudesmanoids are particularly widespread, and have special trends of accumulation in particular Asteraceae tribes and subtribes.⁵⁹³

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