

REVIEW

The Chemistry on Diterpenoids in 1980

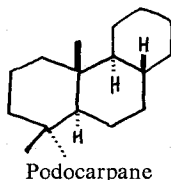
Eiichi FUJITA*, Kaoru FUJI, Yoshimitu NAGAO, Manabu NODE,
and Masahito OCHIAI

Received April 18, 1983

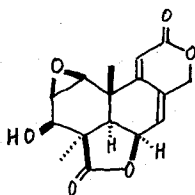
I. INTRODUCTION

This is one of a series of our annual reviews on diterpenoid chemistry. The style is changed from that in the previous reviews. The following abbreviations are used. [CN]: common name; [NS]: natural source; [REF]: reference number; [NC]: notes and comments.

II. PODOCARPANE DERIVATIVES

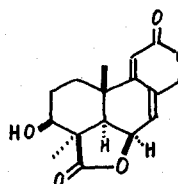


1) Isolation and Structure Determination



1

[CN] wentilactone A
[NS] *Aspergillus wentii*
[REF] 1
[NC] X-ray analysis

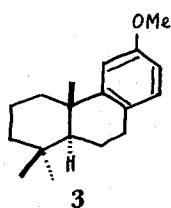


2

[CN] wentilactone B
[NS] *Aspergillus wentii*
[REF] 1

* 藤田榮一, 富士 薫, 長尾善光, 野出 学, 落合正仁: Cancer Drug Research Laboratory, Institute for Chemical Research, Kyoto University, Uji, Kyoto 611.

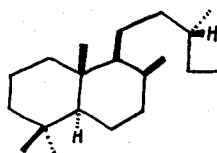
2) **Synthesis and Reaction**



[REF] 2

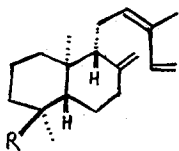
[NC] biogenetic-type cyclization

III. LABDANE DERIVATIVES



Labdane

1) **Isolation and Structure Determination**



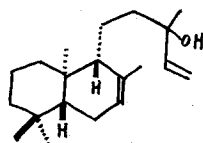
4 R=Me

5 R=CHO

6 R=CO₂H

[NS] *Helianthus* species

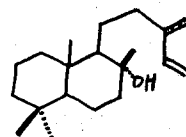
[REF] 3



7

[NS] *Helichrysum* species

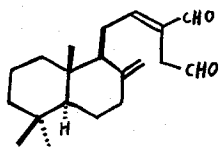
[REF] 4



8

[NS] *Smallanthus fruticosus*

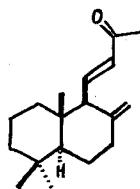
[REF] 5



9

[NS] *Alpinia speciosa*

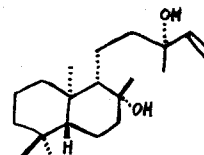
[REF] 6



10

[NS] *Alpinia speciosa*

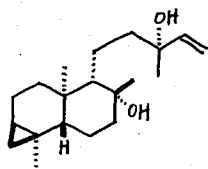
[REF] 6



11

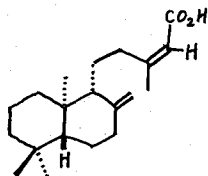
[NS] *Gnaphalium undulatum*

[REF] 7



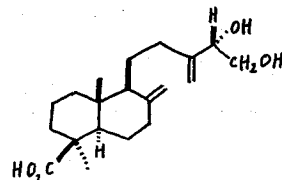
12

[NS] *Gnaphalium undulatum*
[REF] 7



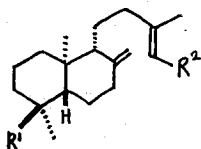
13

[NS] *Morithamnus crassus*
[REF] 8



14

[NS] *Juniperus communis*
[REF] 9



15 $R^1 = \text{CO}_2\text{H}$, $R^2 = \text{CH}_2\text{OH}$

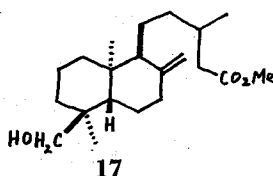
[CN] visidic acid A

16 $R^1 = \text{CO}_2\text{H}$, $R^2 = \text{CH}_2\text{OAc}$

[CN] visidic acid B

[NS] *Chrysothamnus viscidiflorus*

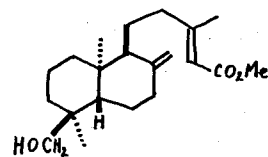
[REF] 10



17

[NS] rosin of
"Brazil capal"

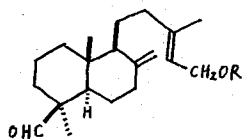
[REF] 11



18

[NS] rosin of
"Brazil capal"

[REF] 11

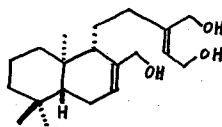


19 $R = \text{H}$

20 $R = \text{D-xylose}$

[NS] *Thujopsis dolabrata*

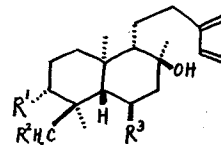
[REF] 12



21

[NS] *Achyrocline alata*

[REF] 13



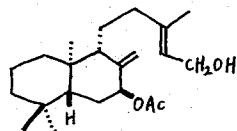
22 $R^1 = \text{H}$, $R^2 = \text{OH}$, $R^3 = \text{OAc}$

23 $R^1 = \text{H}$, $R^2 = \text{OAc}$, $R^3 = \text{OH}$

24 $R^1 = \text{OH}$, $R^2 = \text{H}$, $R^3 = \text{OAc}$

[NS] *Sideritis foetens*

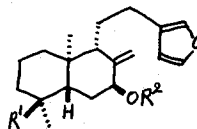
[REF] 14



25

[NS] *Austroeupeatorium*
chaparensis

[REF] 15



26 $R^1 = \text{Me}$, $R^2 = \text{Ac}$

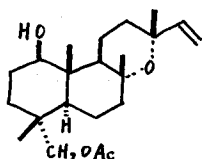
27 $R^1 = \text{CHO}$, $R^2 = \text{Ac}$

[NS] *Austroeupeatorium chaparensis*

[REF] 15

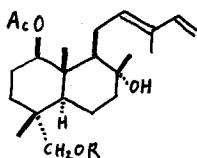
28 $R^1 = \text{CH}_2\text{OAc}$, $R^2 = \text{Ac}$

29 $R^1 = \text{CH}_2\text{OH}$, $R^2 = \text{Ac}$



30

[NS] *Aristequietia
buddleaefolia*
[REF] 16



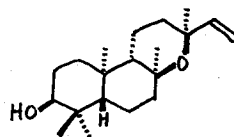
31 R=CO(CH₂)₁₈Me

32 R=COCH=CHC₆H₄OH(*p*)

33 R=COCH=CHC₆H₄OAc(*p*)

[NS] *Aristequietia buddleaefolia*

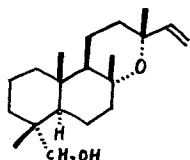
[REF] 16



34

[NS] *Subtribus espletinae*

[REF] 17

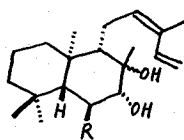


35

[CN] jhanol

[NS] *Stevia rebaudiana*

[REF] 18



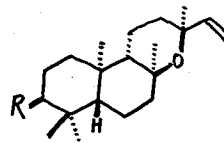
36 R=OH

[CN] austroinulin

37 R=OAc

[NS] *Stevia rebaudiana*

[REF] 18

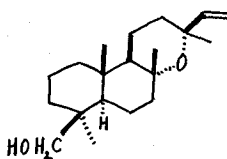


34 R=OH

38 R=H

[NS] *Libanothamnus spectabilis*

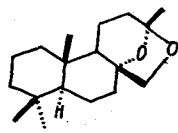
[REF] 19



39

[NS] *Baccharis tola*

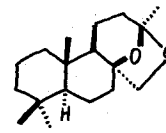
[REF] 20



40

[NS] *Pinus monticola*

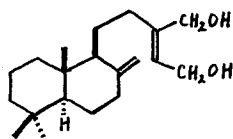
[REF] 21



41

[NS] *Pinus monticola*

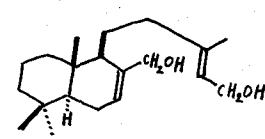
[REF] 21



42

[NS] *Ceroplastes ceriferus*

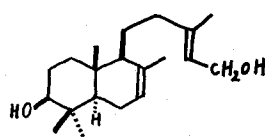
[REF] 22



43

[NS] *Ceroplastes ceriferus*

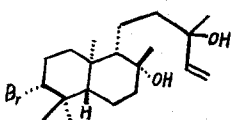
[REF] 22



44

[NS] *Ceroplastes ceriferus*

[REF] 22

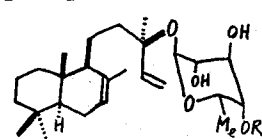


45

[CN] isoconcinndiol

[NS] *Laurencia snyderae*
var. guadalupensis

[REF] 23

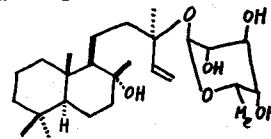


46 R=H

47 R=Ac

[NS] *Aster spathulifolius*

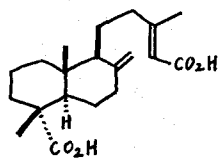
[REF] 24



48

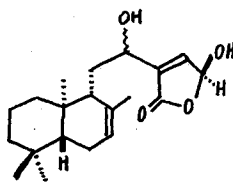
[NS] *Aster spathulifolius*

[REF] 24



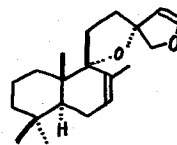
49

[CN] dehydropinifolic acid
[NS] *Pinus silvestris*
[REF] 25



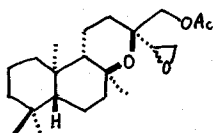
50

[NS] *Acritopappus* species
[REF] 26



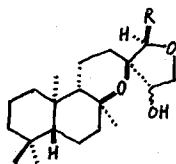
51

[NS] *Solidago* species
[REF] 27



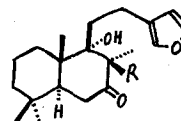
52

[NS] *Schkuhria* species
[REF] 28



53 R=OH

54 R=OAc
[NS] *Schkuhria* species
[REF] 28

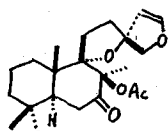


55 R=H

[CN] hispanolone

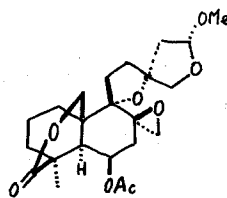
56 R=OAc

[CN] galeopsin
[NS] *Galeopsis angustifolia*
[REF] 29



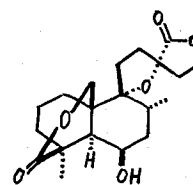
57

[CN] pregaleopsin
[NS] *Galeopsis angustifolia*
[REF] 29



58

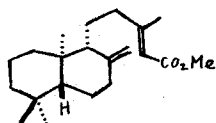
[CN] methoxynepetaefolin
[NS] *Leonotis nepetaefolia*
[REF] 30
[NC] X-ray analysis



59

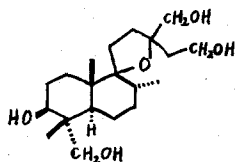
[CN] nepetaefolinol
[NS] *Leonotis nepetaefolia*
[REF] 30

2) Synthesis and Reaction



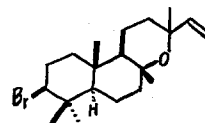
60

[CN] methyl copalate
[REF] 31
[NC] synthesis of
debromoisoaplysin-20
and its C-13 epimer
from 60



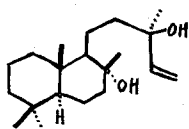
61

[CN] lagochilin
[REF] 32
[NC] acetylation of 61
NMR studies



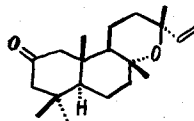
62

[REF] 33
[NC] brominative
cyclization of geranyl-
linalool



63

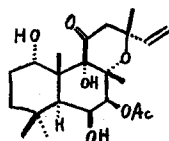
- [CN] sclareol
 [REF] 34~37
 [NC] oxidation of **63** by a chromium mixture



64

- [CN] 2-oxomanoyl oxide
 [REF] 38
 [NC] chemical conversion

3) Miscellaneous Section



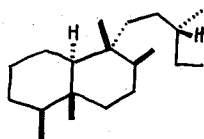
65

- [CN] forskolin
 [NS] *Coleus* and *Plectranthus* species
 [REF] 39
 [NC] TLC and GLC assay methods for the presence of **65**

Additional references

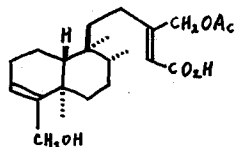
- [REF] 40
 [NC] Inheritance of labdanoid producing ability in *Nicotiana Tabacum*.
 [REF] 41
 [NC] A review article on the chemistry of the *Compositae* diterpenoids.

IV. CLERODANE DERIVATIVES



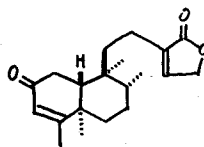
Clerodane

1) Isolation and Structure Determination



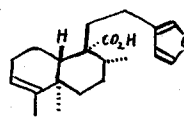
66

- [NS] *Acritopappus* species
 [REF] 26



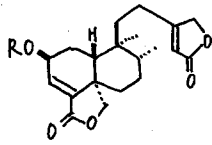
67

- [NS] *Acritopappus* species
 [REF] 26

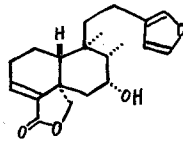


68

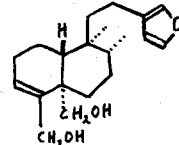
- [NS] *Solidago* species
 [REF] 27



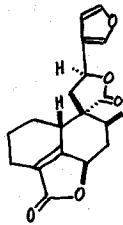
69 R=H
[CN] articulin
70 R=AC
[CN] articulinacetate
[NS] *Baccharis articulata*
[REF] 42



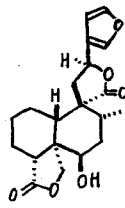
71
[CN] 1-deoxybacrispin
[NS] *Baccharis crispa*
[REF] 43



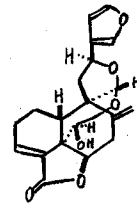
72
[CN] hautriwaic acid
[NS] *Baccharis crispa*
[REF] 43



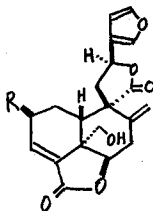
[CN] teuchamaedryn A
[NS] *Teucrium chamaedrys*
[REF] 44



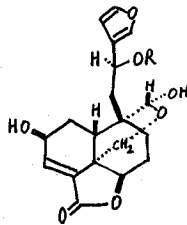
[CN] teuchamaedryn B
[NS] *Teucrium chamaedrys*
[REF] 44



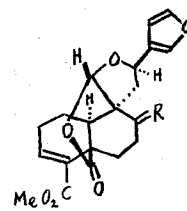
[CN] Plaunol A
[NS] *Croton sublyratus*
[REF] 45
[NC] X-ray analysis



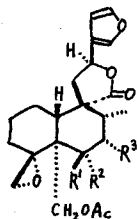
76 R=H
77 R=OH
[CN] plaunol C
[NS] *Croton sublyratus*
[REF] 45



78 R=H
[CN] plaunol D
79 R=Ac
[CN] plaunol E
[NS] *Croton sublyratus*
[REF] 45

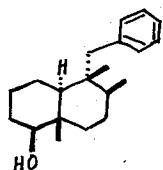


80 R= β -Me, α -H
[CN] dihydrocroverin
[NC] anti-peptic ulcer activity
81 R=CH₂
[CN] croverin
[NS] *Croton verreauxii*
[REF] 46



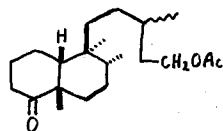
82 R¹, R²=O, R³=OAc
[CN] capitatin
83 R¹=OAc, R²=H, R³=OH
[CN] teucapitatin
[NS] *Teucrium capitatum*
[REF] 47

2) Synthesis and Reaction



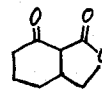
84

[REF] 48
[NC] synthesis



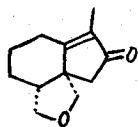
85

[REF] 49
[NC] photoisomerization



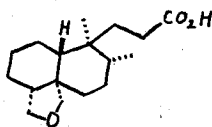
86

[REF] 50
[NC] synthesis



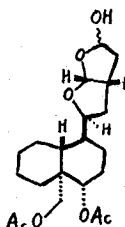
87

[REF] 51
[NC] synthesis



88

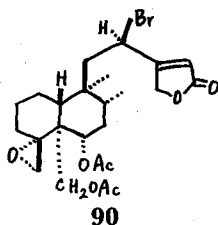
[REF] 52
[NC] synthesis



89

[REF] 53
[NC] synthesis

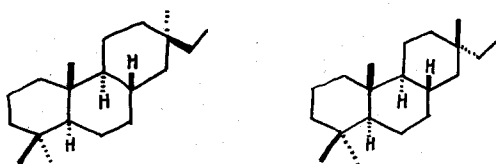
3) Miscellaneous Section



90

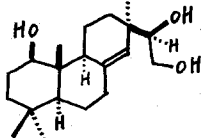
[CN] 12(R)-bromoajugarin-I
[REF] 54
[NC] bromination ·X-ray analysis

V. PIMARANE AND ISOPIMARANE DERIVATIVES



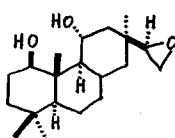
Pimarane and Isopimarane

1) Isolation and Structure Determination



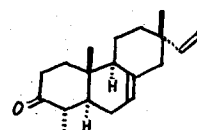
91

[CN] leucophleol
[NS] *Acacia leucophloea*
[REF] 55



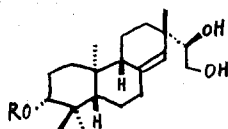
92

[CN] leucophleoxol
[NS] *Acacia leucophloea*
[REF] 55



93

[NS] *Acremonium luzulae*
[REF] 56



94 R=H

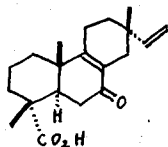
[CN] darutigenol

95 R=β-D-glucose

[CN] darutoside

[NS] *Sigesbeckia orientalis*

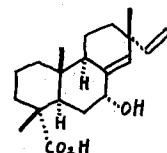
[REF] 57



96

[NS] *Juniperus communis*

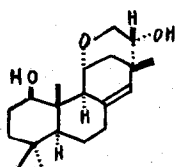
[REF] 9



97

[NS] *Juniperus communis*

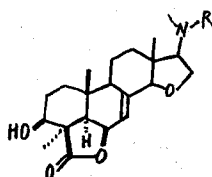
[REF] 9



98

[NS] *Acacia leucophloea*

[REF] 58



99 R=Me

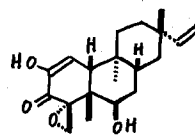
[CN] icacine

100 R=H

[CN] de-N-methylcaceine

[NS] *Icacina guesfeldtii*

[REF] 59

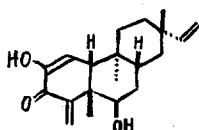


101

[CN] oxidopanamensin

[NS] *Rondeletia panamensis*

[REF] 60



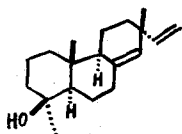
102

[CN] panamensin

[NS] *Rondeletia panamensis*

[REF] 60

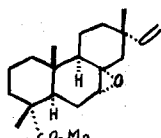
2) Synthesis and Reaction



103

[REF] 61

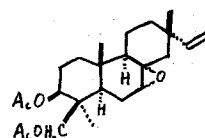
[NC] synthesis of **103**
and its C-4 epimer



104

[REF] 62

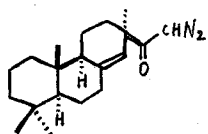
[NC] rearrangement of
104 with $\text{BF}_3\text{-Et}_2\text{O}$



105

[REF] 63

[NC] acid catalyzed ring
cleavage of **105**

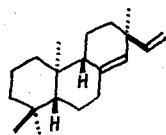


106

[REF] 64

[NC] acid catalyzed cyclization of 106

3) Miscellaneous Section



107

[CN] *ent*-sandaracopimaradiene

[REF] 65

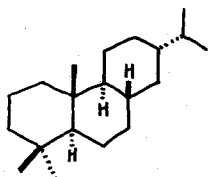
[NC] biosynthesis

Additional references

[REF] 66-68

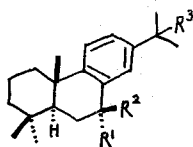
[NC] ¹³C NMR studies

VI. ABIETANE DERIVATIVES



Abietane

1) Isolation and Structure Determination



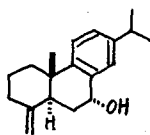
108 R¹=R²=H, R³=OH

109 R¹=OH, R²=R³=H

110 R¹, R²=O, R³=H

[NS] *Pinus monticola*

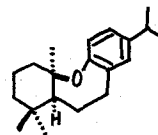
[REF] 21



111

[NS] *Pinus monticola*

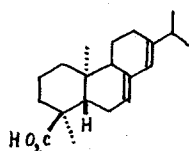
[REF] 21



112

[NS] *Pinus monticola*

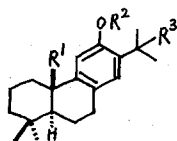
[REF] 21



113

[NS] *solidago* species

[REF] 27



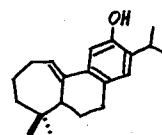
114 R¹=Me, R²=H, R³=OH

115 R¹=CO₂H, R²=Me, R³=H

116 R¹=CHO, R²=R³=H

[NS] *Chamaecyparis pisifera*

[REF] 69

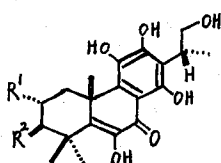


117

[CN] pisiferin

[NS] *Chamaecyparis pisifera*

[REF] 69



118 R¹=R²=H

[CN] coleon C

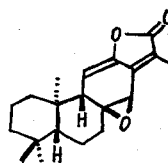
119 R¹=H, R²=OAc

[CN] coleon H

120 R¹=OCHO, R²=H

[NS] *Solenostemon monostachys*

[REF] 70



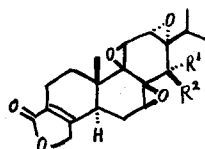
121

[CN] jolkinolide A

[NS] *Euphorbia huachangana*

[REF] 71

2) Synthesis and Reaction



122 R¹=H, R²=OH

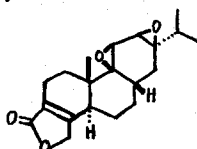
[CN] triptolide

123 R¹, R²=O

[CN] triptonide

[REF] 72, 73

[NS] synthesis

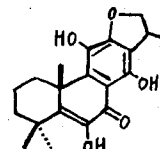


124

[CN] stemolide

[REF] 74

[NC] synthesis

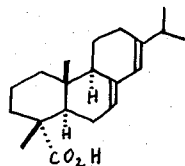


125

[CN] lycoxanthol

[REF] 75

[NC] synthetic studies of **125**

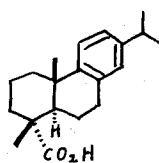


126

[CN] abietic acid

[REF] 76

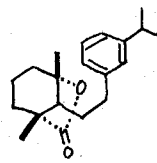
[NC] Formation of coeon A skeleton from **126**



127

[REF] 77

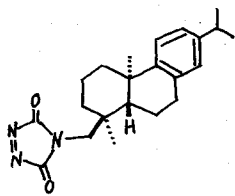
[NC] synthesis of carotene derivative from **127**



128

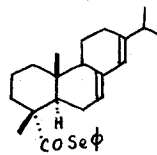
[REF] 78

[NC] synthesis and structure determination



129

[REF] 79
[NC] asymmetric Diels-Alder
reaction of **129**



130

[REF] 80
[NC] reduction of **130** with Bu_3SnH

3) Miscellaneous Section

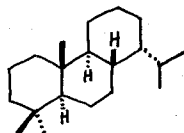
126

[REF] 81
[NC] allergic constituents
of some plants

Additional reference

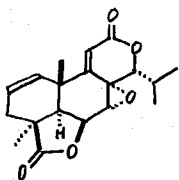
[REF] 82
[NC] studies of hypocholesterolemic
activity of abietamide derivatives

VII. TOTARANE DERIVATIVES



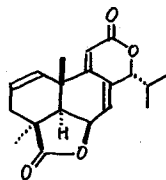
Totarane

1) Isolation and Structure Determination



131

[CN] milanjilactone A
[NS] *Podocarpus milanjanus*
[REF] 83



132

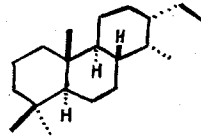
[CN] milanjilactone B
[NS] *podocarpus milanjanus*
[REF] 83

2) Miscellaneous Section

Additional reference

[REF] 84
[NC] ^{13}C NMR studies

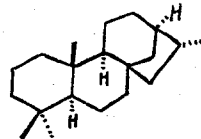
VIII. CASSANE DERIVATIVES



Cassane

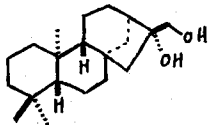
No report

IX. KAURANE DERIVATIVES



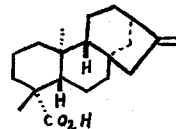
Kaurane

1) Isolation and Structure Determination



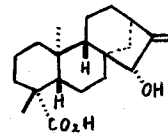
133

[NS] *Morithamnus crassus*
[REF] 8



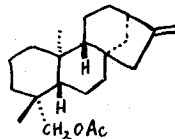
134

[NS] *Helianthus niveus* and
others
[REF] 3, 4, 7, 19, 85, 86, 87



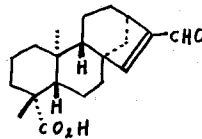
135

[NS] *Helianthus niveus*
and others
[REF] 7, 85; cf. 86



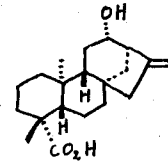
136

[NS] *Espeletiopsis* species
[REF] 88



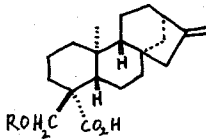
137

[NS] *Espeletiopsis* species
[REF] 88



138

[NS] *Smallanthus wedalis*
[REF] 87

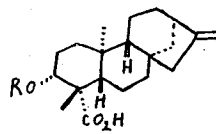


139 R=angeloyl

140 R=seneconyl

141 R=isovaleryl

[NS] *Smallanthus wedalis*
[REF] 87

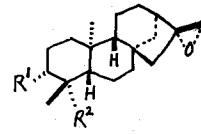


142 R=angeloyl

143 R=seneconyl

144 R=isovaleryl

[NS] *Smallanthus wedalis*
[REF] 87

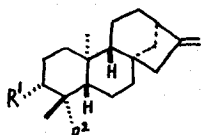


145 R¹=H, R²=CH₂O-
angeloyl

146 R¹=O-angeloyl,
R²=Me

147 R¹=O-angeloyl,
R²=CHO

[NS] *Smallanthus wedalis*
[REF] 87



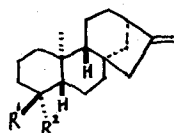
148 R¹=OAc, R²=CO₂H

149 R¹=OH, R²=CH₂OH

150 R¹=OH, R²=CO₂H

[NS] *Stachys lanata*

[REF] 89



151 R¹=R²=Me

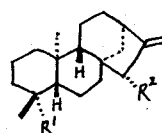
152 R¹=Me, R²=H

153 R¹=H, R²=Me

[NS] *Libanothamnus*

granatesianus

[REF] 19



154 R¹=CO₂H, R²=OAc

155 R¹=CO₂H, R²=O

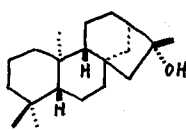
isovaleryl

156 R¹=CH₂OH, R²=H

[NS] *Libanothamnus*

granatesianus

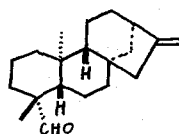
[REF] 19



157

[NS] *Helichrysum* species

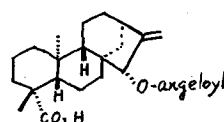
[REF] 4



158

[NS] *Gnaphalium undulatum*

[REF] 7

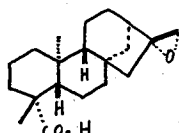


159

[NS] *Smallanthus fruticosus*

and others

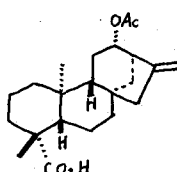
[REF] 3, 5, 27



160

[NS] *Smallanthus fruticosus*

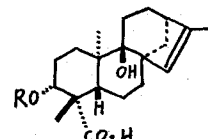
[REF] 5



161

[NS] *Helianthus* species

[REF] 3

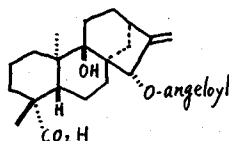


162 R=isovaleryl

163 R=isobutyl

[NS] *Polymnia canadensis*

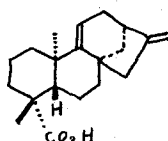
[REF] 90



164

[NS] *Steiractinia mollis*

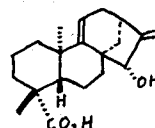
[REF] 91



165

[NS] *Steiractinia mollis* and others

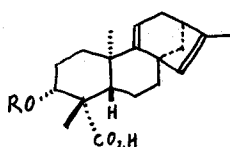
[REF] 3, 7, 19, 91, 92



166

[NS] *Smallanthus fruticosus*

[REF] 5



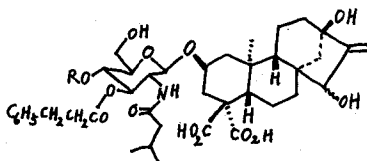
167 R=isovaleryl

168 R=isobutyl

169 R=tigl

[NS] *Polymnia canadensis*

[REF] 90



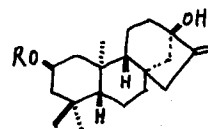
170 R=H

[CN] wedeloid

171 R=L-rhamnopyranosyl

[NS] *Wedelia asperrima*

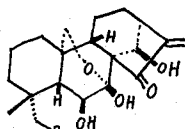
[REF] 93



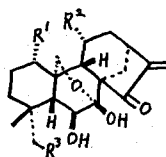
172 R=β-D-glucosyl

[NS] *Lindsaea chienii*

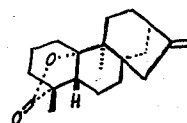
[REF] 94



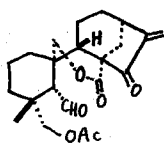
- 173** R=H
174 R=OAc
 [CN] longikaurin A (173)
 and B (174)
 [NS] *Rabdosia longituba*
 [REF] 95



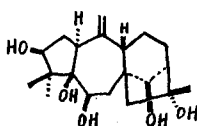
- 175** R¹=OH, R²=R³=H
176 R¹=OAc, R²=R³=H
177 R¹=OH, R²=H,
 R³=OAc
178 R¹=R³=OAc, R²=H
179 R¹=R²=OH, R³=H
 [CN] effusanin A-E
 [NS] *Rabdosia effusa*
 [REF] 96



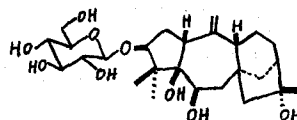
- 190**
 [CN] tetrachyryrin
 [NS] *Tetrachyryrin*
orizabaensis
 [REF] 97



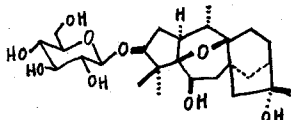
- 191**
 [CN] effusin
 [NS] *Rabdosia effusus*
 [REF] 98



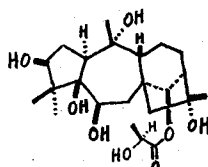
- 192**
 [CN] grayanotoxin II
 [REF] 99



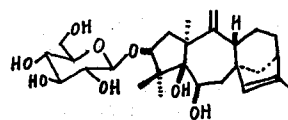
- 193**
 [CN] grayanoside C
 [NS] *Leucothoe grayana*
 [REF] 100



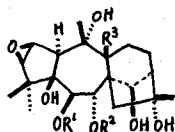
- 194**
 [CN] grayanoside D
 [NS] *Leucothoe grayana*
 [REF] 101



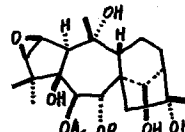
- 195**
 [CN] asebotoxin-X
 [NS] *Pieris japonica*
 [REF] 102, 103



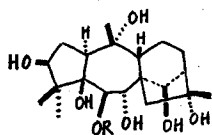
- 196**
 [CN] pieroside A
 [NS] *Pieris japonica*
 [REF] 102



- 197** R¹=R²=R³=H
198 R¹=Ac, R²=H, R³=OH
199 R¹=Ac, R²=COC₂H₅, R³=OH
 [CN] pieristorin H, J, K
 [NS] *Pieris japonica*
 [REF] 103



- 200** R=H
201 R=Ac
 [CN] kalmixitoxin IV, V
 [NS] *kalmia latifolia*
 [REF] 104



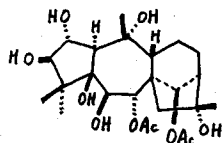
202 R=H

203 R=Ac

[CN] kalmitoxin I, III

[NS] *Kalmia latifolia*

[REF] 104

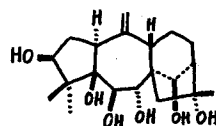


204

[CN] kalmitoxin VI

[NS] *Kalmia latifolia*

[REF] 104



205

[CN] kalmitoxin II

[NS] *Kalmia latifolia*

[REF] 104

Additional references

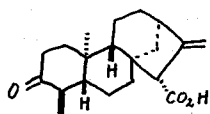
[REF] 105

[NC] studies of constituents of *Sideritis flavoviren*

[REF] 106

[NC] studies of constituents of *Eupatorium tinofolium*

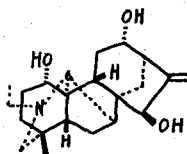
2) Synthesis and Reaction



206

[REF] 107

[NC] synthesis of optically active **206**

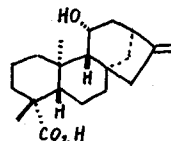


207

[CN] napelline

[REF] 108

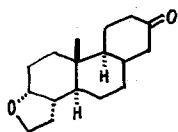
[NC] total synthesis



208

[REF] 109

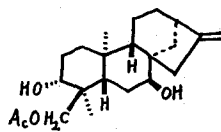
[NC] conversion of **165** to **208** and 12-hydroxy derivatives



209

[REF] 110

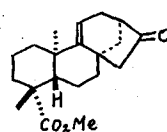
[NC] intermediate to epoxynorcafestanone



210

[REF] 111

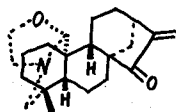
[NC] oxidation with SeO₂-H₂O₂



211

[REF] 112

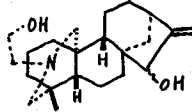
[NC] 10α→9α-methyl migration



212

[REF] 113

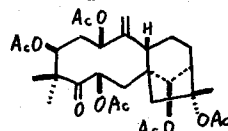
[NC] alumina catalyzed addition of amine to **212**



213

[REF] 114

[NC] oxidative cyclization

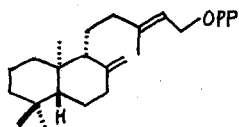


214

[REF] 115

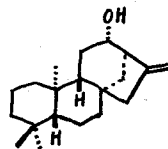
[NC] conversion of grayanotoxin II tetraacetate to **214**

3) **Miscellaneous Section**



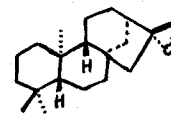
215

[CN] copalyl pyrophosphate
[REF] 116, 117
[NC] enzymatic cyclization
of **215** to *ent*-kaurene



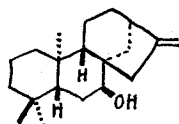
216

[REF] 118
[NC] microbial transfor-
mation of **216**



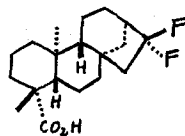
217

[REF] 119
[NC] inhibition of gib-
berellic acid bi-
osynthesis by **217**



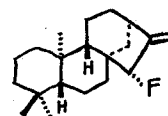
218

[REF] 120
[NC] microbial transformation
of diterpenoids



219

[REF] 121
[NC] microbial
transformation



220

[REF] 122
[NC] Biological activity
of some fluorodi-
terpenoids

Additional references

[REF] 123, 124

[NC] ¹³C NMR studies

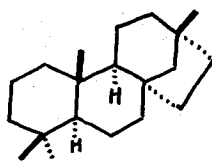
[REF] 125

[NC] analytical studies of
stevioside of *stevia*
rebaudiana

[REF] 126

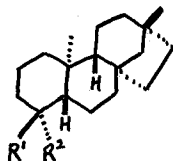
[NC] A review article on
diterpenoid alkaloids

X. BEYERANE DERIVATIVES

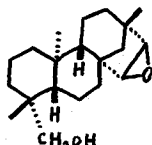


Beyerane

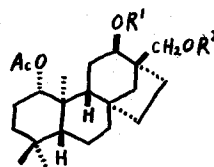
1) Isolation and Structure Determination



221 $R^1=Me, R^2=CH_2OH$
 [CN] erythroxylyol-A
222 $R^1=CH_2OH, R^2=Me$
 [NS] *Baccharis tola*
 [REF] 20

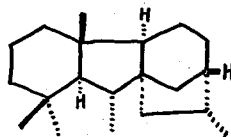


223
 [NS] *Baccharis tola*
 [REF] 20



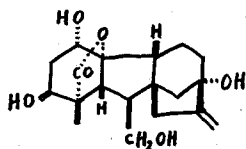
224 $R^1=H, R^2=Ac$
225 $R^1=Ac, R^2=H$
 [NS] *Sideritis serrata*
 [REF] 127

XI. GIBBERELLANE DERIVATIVES



Gibberellane

1) Isolation and Structure Determination

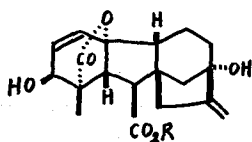


226
 [CN] gibberellin A_{57}
 [NS] *Gibberilla fujikuroi*
 [REF] 128

Additional references

- [REF] 129
- [NC] identification of gibberellins in Spinach shoots
- [REF] 130
- [NC] identification of gibberellins in wheat grain
- [REF] 131
- [NC] gibberellins in mature apple seeds
- [NS] *pharbitis purpurea*
- [REF] 132
- [NC] glucosyl esters of GA_5 and A_{44}

2) Synthesis and Reaction

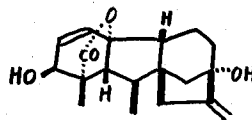


227 R=H

- [CN] gibberellin A₃
- [REF] 133, 134
- [NC] total synthesis
- [REF] 135
- [NC] introduction of fluorine at C-4
- [REF] 136
- [NC] rearrangement
- [REF] 137
- [NC] Pd (OAc)₂-catalyzed reactions
- [REF] 138
- [NC] partial synthesis of 16, 17-dihydro-*D*¹⁵-G A₃

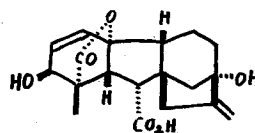
228 R=CH₃

- [REF] 139
- [NC] photochemistry, crystal structure
- [REF] 140
- [NC] introduction of chlorine and bromine at C-7
- [REF] 141
- [NC] 3-O-acetyl derivative, reaction with PBr₃



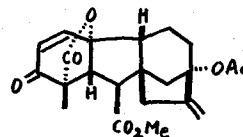
229

- [REF] 142
- [NC] chemical conversion from GA₃



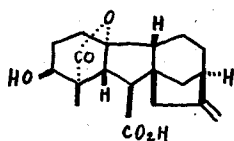
230

- [CN] 6-*epi*-gibberellin A₃
- [REF] 143
- [NC] chemical conversion from GA₄



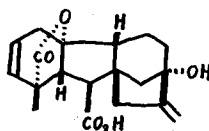
231

- [REF] 144
- [NC] reduction with NaBH₄ or NaBD₄



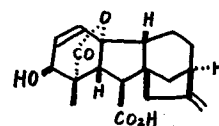
232

- [CN] gibberellin A₄
- [REF] 145
- [NC] total synthesis
- [REF] 146
- [NC] physiological studies
- [REF] 147
- [NC] deuterium labeling



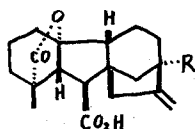
233

- [CN] gibberellin A₅
- [REF] 148
- [NC] 1-deuterated derivative
- [REF] 149
- [NC] partial synthesis, 3-deuterated derivatives



234

- [CN] gibberellin A₇
- [REF] 150
- [NC] partial synthesis



235 R=H

[CN] gibberellin A₉

[REF] 147

[NC] deuterium labeling

[REF] 149

[NC] partial synthesis, 3-deuterated derivative

236 R=OH

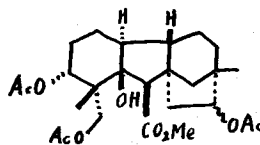
[CN] gibberellin A₂₀

[REF] 147

[NC] deuterium labeling

[REF] 149

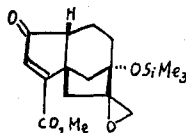
[NC] partial synthesis 3-deuterated derivative



237

[REF] 151

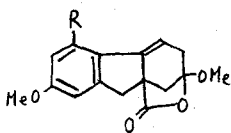
[NC] synthesis



238

[REF] 152

[NC] synthesis

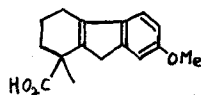


239 R=H

240 R=OMe

[REF] 153

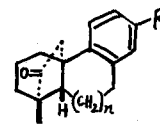
[NC] synthesis



241

[REF] 154

[NC] synthesis

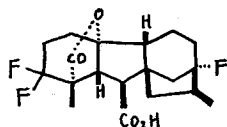


242 n=1 or 2, R=H

243 n=1 or 2, R=OMe

[REF] 155

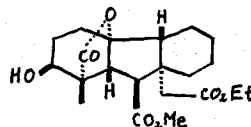
[NC] synthesis



244

[REF] 156

[NC] chemical conversion from benzyl gibberellate

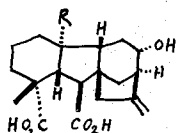


245

[REF] 157

[NC] photoproduct from gibberellin C, X-ray analysis

3) Miscellaneous Section

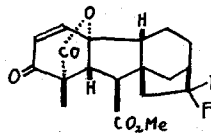


250 R=Me

251 R=CO₂H

[REF] 118

[NC] microbial transformations



252

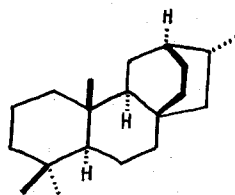
[REF] 121

[NC] microbial transformations

Additional references

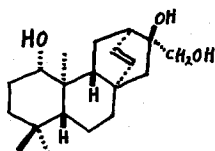
- | | | |
|--|--------------------------------------|---------------------------------|
| [REF] 158 | [REF] 160 | [REF] 162 |
| [NC] application of gibberellic acid in treatment of lung cancer | [NC] HPLC separation of gibberellins | [NC] a review on plant hormone |
| [REF] 159 | [REF] 161 | [REF] 163 |
| [NC] phytohormone activity of gibberellin A ₃ | [NC] mass spectrometric studies | [NC] metabolism of gibberellins |

XII. ATISANE DERIVATIVES



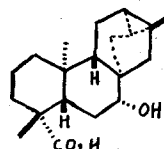
Atisane

1) Isolation and Structure Determination



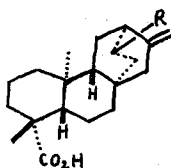
253

- [CN] sideritol
 [REF] 164
 [NC] X-ray analysis of *p*-bromobenzoate

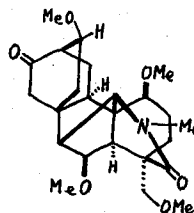


254

- [CN] ciliaric acid
 [NS] *Helianthus niveus*
 [REF] 85

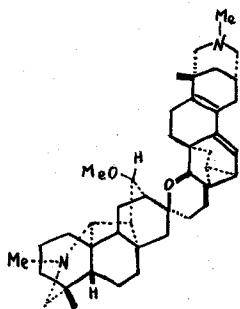


- 255 R=O-ang
 256 R=O-isoval
 [NS] *Helianthus* species
 [REF] 3



257

- [REF] 165
 [NS] X-ray analysis, intermediate for total synthesis



258

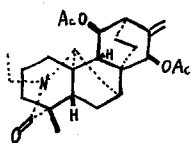
[CN] staphisine

[NS] *Delphinium staphisagria*

[REF] 166

[NC] X-ray analysis

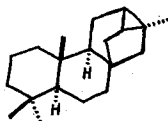
2) Synthesis and Reaction



259

[REF] 167

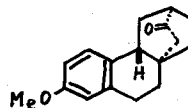
[NC] total synthesis



260

[REF] 168

[NC] chemical conversion



261

[REF] 169

[NC] synthesis

3) Miscellaneous Section

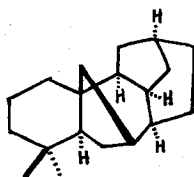
[REF] 41

[NC] a review

[REF] 126

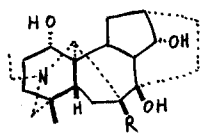
[NC] a review

XIII. ACONANE DERIVATIVES



Aconane

1) Isolation and Structure Determination



262 R=H

[CN] cardiopetaline

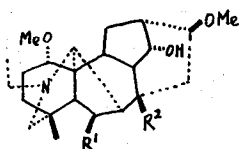
263 R=OH

[CN] cardiopetalidine

[NS] *Delphinium cardiopetalum*

[REF] 170

[NC] X-ray analysis of 262



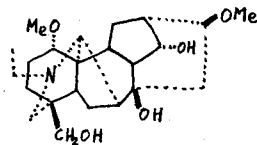
264 R¹=OAc, R²=OMe

265 R¹=R²=OH

[NS] *Delphinium bicolor*

[REF] 171

[NC] X-ray analysis



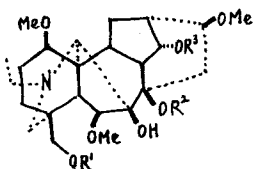
266

[CN] cammaconine

[NS] *Aconitum variegatum*

[REF] 172

[NC] structure revision



267 R¹=R²=Me, R³=Ac

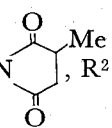
[CN] ambigaine

268 R¹=Me, R²=H, R³=Ac

[CN] 14-acetylbrowniine

269 R¹=COC₆H₄-*o*-NHAc, R²=H, R³=Ac

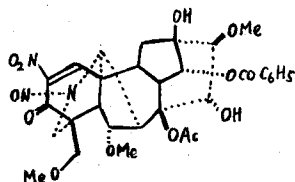
[CN] ajadine

270 R¹=COC₆H₄-*o*-N , R²=H, R³=Bz

[CN] ajacusine

[NS] *Consolida ambigua*

[REF] 173



271

[REF] 174

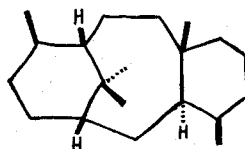
[NC] oxidation product of
aconitine

Additional reference

[REF] 126

[NC] a review

XIV. TAXANE DERIVATIVES



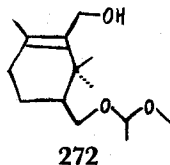
Taxane

1) Isolation and Structure Determination

[REF] 175

[NC] a review

2) Synthesis and Reaction

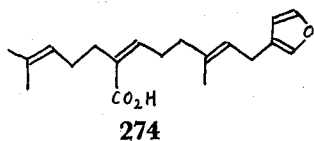
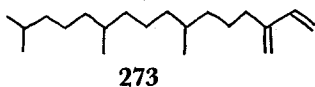


[REF] 176

[NC] synthesis of a key intermediate

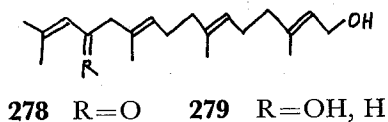
XV. THE OTHERS

1) Isolation and Structure Determination



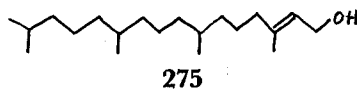
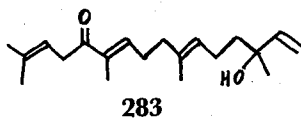
[CN] neophytadiene (273)
centipedic acid (274)

[NS] *Plagiocheilus prostratus*
[REF] 177

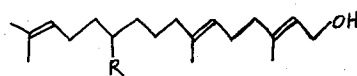
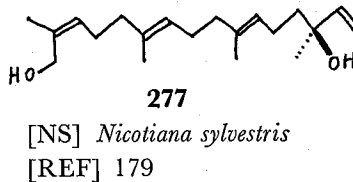
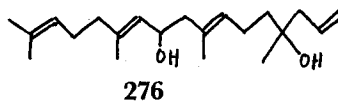


[CN] eleganolone (278), elegandiol (279)
epoxy eleganolone (280)

[NS] *Cystoseira elegans* and *Bifurcaria bifurcata*
[REF] 180 and 181



[CN] phytol
[NS] *Thymelea hirsuta*
[REF] 178

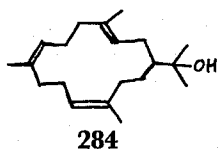


282 R=CHO

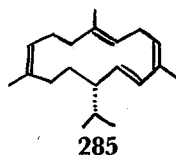
[NS] *Croton kerrii*
[REF] 182

[REF] 180

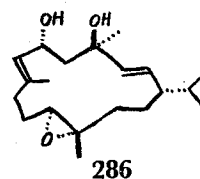
[NC] acyclic diterpenes from
mediterranean cystoseiraceae



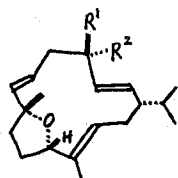
[NS] *Helichrysum* species
[REF] 4



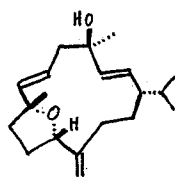
[CN] cembrene
[NS] *Nicotiana tabacum*
[REF] 183



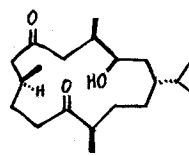
[NS] Greek tobacco
[REF] 184



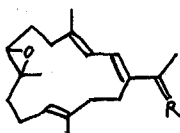
[NS] Greek tobacco
[REF] 184



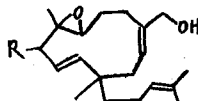
[NS] Greek tobacco
[REF] 184



[CN] plexauralone
[NS] *Plexaura*-related species
[REF] 185
[NC] X-ray analysis

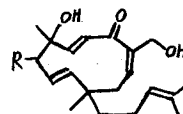


[NS] *Sarcophyton crassocaule*
[REF] 186

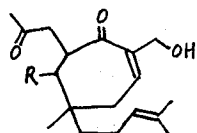


294 $R=H$

[CN] vibsamine A (**293**)
vibasamine B (**295**)
vibasamine F (**294**)



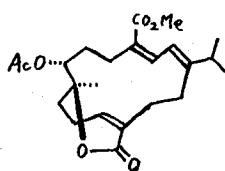
[NS] *Viburnum odoratissimum*
[REF] 187



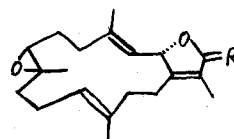
[CN] emblide
[NS] *Sarcophyton glaucum*
[REF] 188

[NC] X-ray analysis

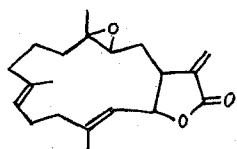
[NS] *Viburnum odoratissimum*
[REF] 187



[CN] deoxosarcophine (**299**)
sarcophine (**300**)
[NS] *Sarcophyton* species
[REF] 189



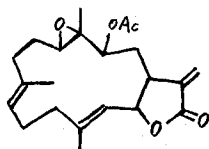
[NS] *Sarcophyton* species
[REF] 189



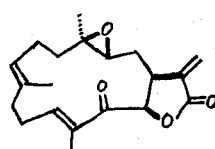
301

[NS] *Lobophytum pauciflorum*

[REF] 190



302



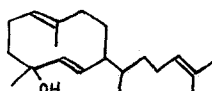
303

[CN] peunicin

[NS] *Eunicea succinea*

[REF] 191

[NC] X-ray analysis

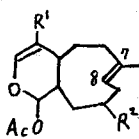


304

[CN] obscuronatin (**304**), xeniculin (**305**)

[NS] *Xenia macrospiculata* and *X. obscuronate*

[REF] 192

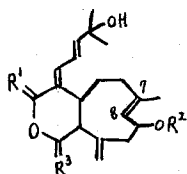


305 R¹=, R²=OAc

306 R¹=, R²=H

307 R¹=, R²=H,

7, 8-epoxide



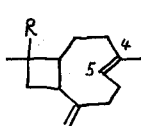
308 R¹=O, R²=H, R³=H₂

309 R¹=H₂, R²=H, R³=O

310 R¹=H₂, R²=Ac, R³=O

311 R¹=O, R²=H, R³=H₂, 7, 8-epoxide

312 R¹=H₂, R²=H, R³=OH, H



313 R=CH₂-

314 R=CH₂-, 4,5-epoxide

315 R=CH₂-

316 R=CH₂-, 4,5-epoxide

317 R=CH₂-

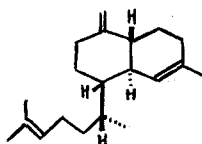
318 R=CH₂-

319 R=CH₂-, 4,5-epoxide

[CN] xeniolide-A (**308**) xeniolide-B (**309**), xenialactol (**312**), xeniaphyllenol (**313**), isoxeniaphyllenol (**315**)

[NS] *Xenia macrospiculata* and *X. obscuronate*

[REF] 192

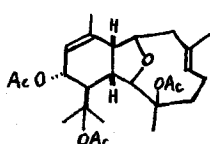


320

[NS] *Cubitermes umbratus*

[REF] 193

[NC] X-ray analysis

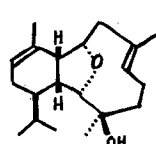


321

[CN] ophirin

[NS] *Muricella* species

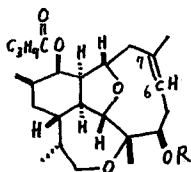
[REF] 194



322

[NS] pacific soft coral

[REF] 195



338 R=Ac, 6, 7-trans

[CN] asbestinin-1

339 R=H, 6, 7-trans

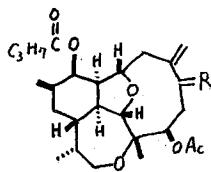
[CN] asbestinin-3

340 R=Ac, 6, 7-cis

[CN] asbestinin-2

[REF] 202

[NC] X-ray analysis (**338**)

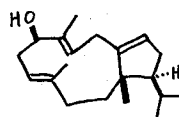


341 R=O

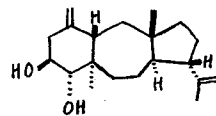
[CN] asbestinin-4

342 R= α -OH, β -H

[CN] asbestinin-5



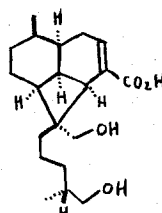
343



344

[NS] *Clavularia inflata*

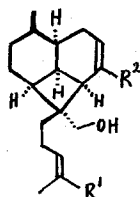
[REF] 203



345

[NS] *Eremophila* species

[REF] 204



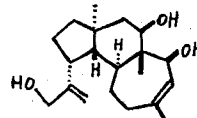
346 R¹=R²=CH₂OH

347 R¹=CO₂H, R²=Me

348 R¹=CO₂H, R²=CH₂OH

[NS] *Eremophila decipiens*

[REF] 205



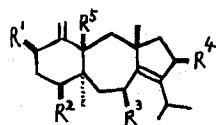
349

[CN] onychiol B

[NS] *Onchium japonicum*

[REF] 206

[NC] X-ray analysis



350 R¹=H, R²=R⁵=OH,

R³=R⁴=OAc

351 R¹=R³=R²=H,

R²=R⁵=OH

352 R¹=R⁵=OH,

R²=R³=R⁴=H

353 R¹=R³=R⁴=R⁵=H,

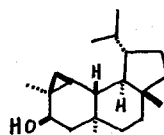
R²=OH

[CN] amijidietylol (**350**), amijiol

(**351**), isoamijiol (**352**)

[NS] *Dictyota linearis*

[REF] 207 and 208



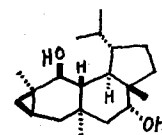
354

[CN] neoverrucosan-5 β -ol

[NS] *Mylia verrucosa*

[REF] 209

[NC] X-ray analysis



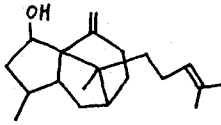
355

[CN] dihydroxy-verrucosane

[NS] *Mylia verrucosa*

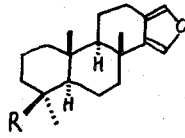
[REF] 210

[NC] X-ray analysis



356

[CN] stoechospermol
[NS] *Stoechospermum marginatum*
[REF] 211

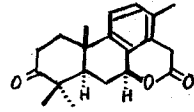


357 R=CO₂H

358 R=CHO

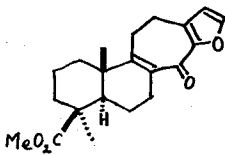
359 R=Me

[NS] *Spongia officinalis*
[REF] 212



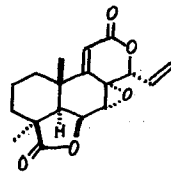
360

[NS] *Vellozia compacta*
[REF] 213



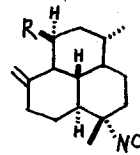
361

[CN] hispanonic acid methyl ester
[NS] *Balloya hispanica*
[REF] 214
[NC] X-ray analysis



362

[CN] Salignone-D
[NS] *Podocarpus saligna*
[REF] 215
[NC] X-ray analysis

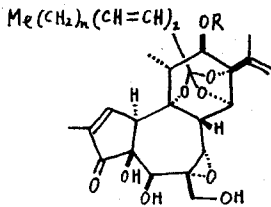


363 R=CH=CMe₂

364 R=CH₂-

365 R=CH₃-NC

[NS] *Adosia* species
[REF] 216



R=CO(CH=CH)₃(CH₂)₂CH₂OY
n=2

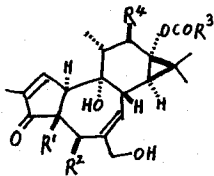
366 Y=H, **367** Y=CO(CH₂)₁₂Me

368 Y=COCH=CH(CH₂)₁₁Me

369 Y=COC₁₃H₂₇

370 Y=COC₁₄H₂₇

[NS] *Stillingia sylvatica* [REF] 217

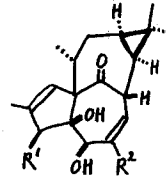


371 R¹=R²=H, R³=CH₂CH Me₂
R⁴=OCOC(Me)=CHMe

[NS] *Synadenium grantii*
[REF] 218

372 R¹=R²=OH, R⁴=H
R³=(CH₂)₁₄Me

[NS] *Stillingia sylvatica*
[REF] 217



373 R¹=angelate, R²=Me

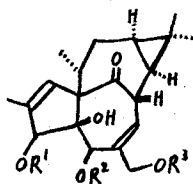
374 R¹=hexanoate, R²=Me

375 R¹=angelate, R²=CH₂OH

[NS] *Euphorbia paralias*

[REF] 219

[NC] Irritant and cytotoxic constituents



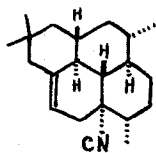
376 $R^1 = \text{COC}_2\text{H}_5$, $R^2 = \text{H}$, $R^3 = \text{COCH}(\text{Me})\text{C}_2\text{H}_5$

377 $R^1 = R^2 = \text{H}$, $R^3 = \text{COCHMe}_2$

378 $R^1 = \text{COC}_2\text{H}_5$, $R^2 = \text{H}$, $R^3 = \text{COCHMe}_2$

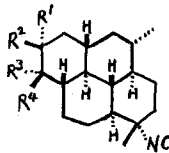
399 $R^1 = R^3 = \text{COCHMe}_2$, $R^2 = \text{H}$

[NS] *Euphorbia cotinifolia* [REF] 220



380

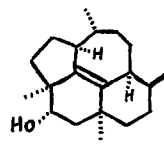
[NS] *Adocia* species



381 $R^1 = \text{NC}$, $R^2 = R^3 = \text{Me}$, $R^4 = \text{H}$

382 $R^1 = R^4 = \text{Me}$, $R^2 = \text{H}$, $R^3 = \text{NC}$

[REF] 216

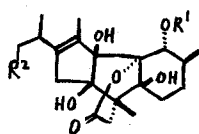


383

[NS] *Nasutitermes rippertii* and *N. ephratae*

[REF] 221

[NC] X-ray analysis

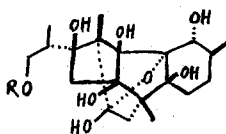


384 $R^1 = \text{Ac}$, $R^2 = \text{H}$

385 $R^1 = R^2 = \text{H}$

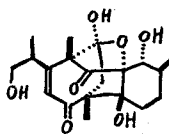
386 $R^1 = \text{H}$, $R^2 = \text{OH}$

387 $R^1 = \text{H}$, $R^2 = \text{O-}\beta\text{-p-glc-pyr}$



388 $R = \text{H}$

389 $R = \beta\text{-D-glc-pyr}$



390

391 $R = \text{H}$

392 $R = \beta\text{-D-glc-pyr}$

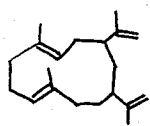
[CN] cinnassiol-A (**386**), -B (**388**), -C (**390**), -D (**391**)

[NS] *Cinnanomum cassia*

[REF] 222, 223, 224, 225

[NC] X-ray analysis

2) Synthesis and Reaction



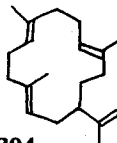
393

[CN] (\pm)-cubitene

[REF] 226

[NC] ·intramolecular coupling of allylic dibromide

·total synthesis



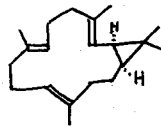
394

[CN] neocembrene

[REF] 227

[NC] ·synthesis of d- and l-neocembrenes and their geometrical isomers

·pheromone activity

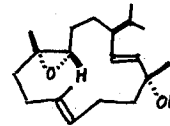


395

[CN] (-)-casbene

[REF] 228

[NC] total synthesis from 1R, 3S(+)-cis-chrystan-themic acid

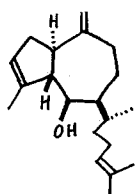


396

[CN] isocembrol (thunbergol)

[REF] 229

[NC] epoxidation



397

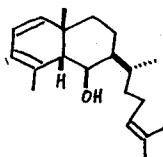
[CN] (+)-pachydietylol A (**397**)

(-)-dietylolene (**398**)

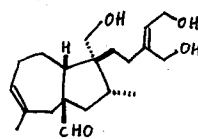
[REF] 230

[NC] total syntheses from

(-)- α -santonin



398



399

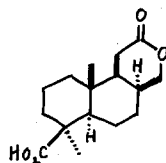
[CN] (\pm)-portulal

[REF] 231

[NC] total synthesis

[REF] 232

[NC] selective degradation of side chain

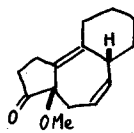


400

[CN] dihydro-8-epi-acrostalidic acid

[REF] 233

[NC] total synthesis

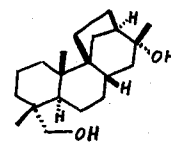


401

[REF] 234

[NC] synthetic approach to phorbol

divinylcyclopropane rearrangement

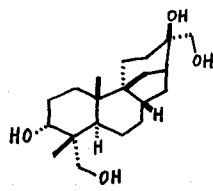


402

[CN] (\pm)-stemarin

[REF] 235

[NC] total synthesis

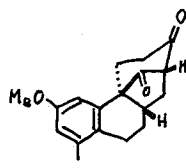


403

[CN] (\pm)-aphidicolin

[REF] 236

[NC] total synthesis

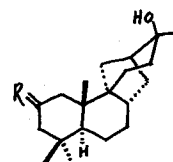


404

[REF] 237

[NC] synthetic approach to aphidicolin

thermolysis of benzocyclobutene



405 R= α -OH, β -H

406 R=O

[CN] (\pm)-stemodin

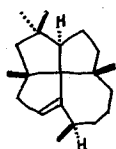
(**405**)

(\pm)-stemodinone

(**406**)

[REF] 238

[NC] total synthesis

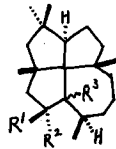


407

[CN] lauren-1-ene

[REF] 239

[NC] osmic acid oxidation
NMR spectra



408 R¹=OH, R²=H, R³= α -H

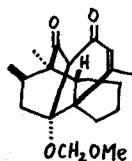
409 R¹=H, R²=OH, R³= α -H

410 R¹=OH, R²=H, R³= β -H

411 R¹=H, R²=OH, R³= β -H

[REF] 240

[NC] remote functionalization with I₂/Pb(OAc)₄

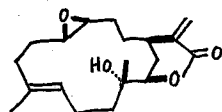


412

[REF] 241

[NC] synthetic precursors to pleuromutilin
one-step synthesis

3) Miscellaneous Section



413

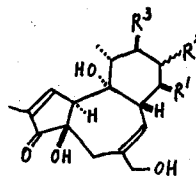
[CN] flexibilide

[REF] 242

[NC] ¹³C-NMR study

spin-lattice

relaxation measurements



414

[REF] 243

[NC] structure-activity relations for
irritant and tumor promoting
activity

tigliane derivatives (414)

$R^1 = H$ or $C(Me)=CH_2$

$R^2 = \alpha$ - or β - $OOC(CH_2)_{12}Me$

$R^3 = H$ or $OOC(CH_2)_6Me$

Additional references

[REF] 244

[NC] structure-activity relationships for phorbol-related diterpene esters

[REF] 41 [NC] a review article on the chemistry of the *Compositae*
(cembrene type diterpenoids)

[REF] 81 [NC] a review article on the plants and plants products that
induced contact dermatitis (tigliane, daphnane, ingenane
type diterpenoids)

[REF] 245 [NC] a review article on the drugs from marine organisms
(cembranoid)

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