

REVIEW

## The Chemistry on Diterpenoids in 1981

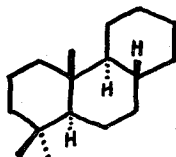
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Manabu NODE\*, and Masahito OCHIAI\*

Received June 1, 1984

### I. INTRODUCTION

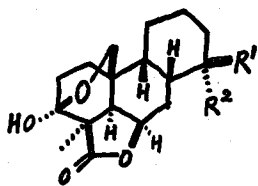
This is one of a series of our annual reviews on diterpenoid chemistry. The following abbreviations are used. [CN]: common name; [NS]: natural source; [REF]; reference number; [NC]: notes and comments.

### II. PODOCARPANE DERIVATIVES



Podocarpane

#### 1) Isolation and Structure Determination



1  $R^1+R^2=0$

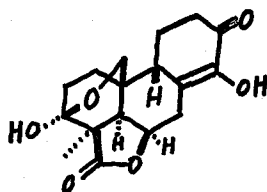
[CN] humirianthenolide A

2  $R^1=H, R^2=OH$

[CN] humirianthenolide B

[NS] *Humirianthera rupestris*

[REF] 1

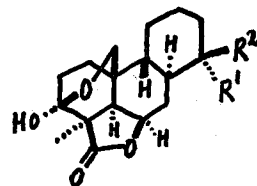


3

[CN] humirianthenolide D

[NS] *Humirianthera rupestris*

[REF] 1



4  $R^1+R^2=0$

[CN] humirianthenolide E

5  $R^1=H, R^2=OH$

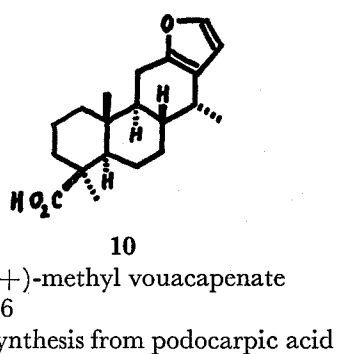
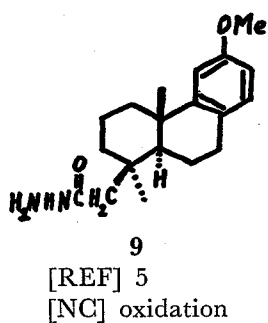
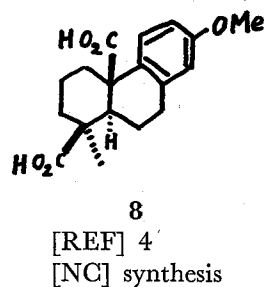
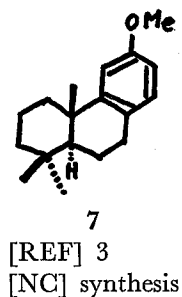
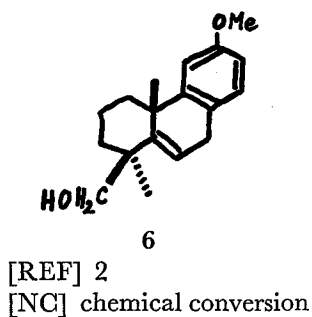
[CN] humirianthenolide F

[NS] *Humirianthera rupestris*

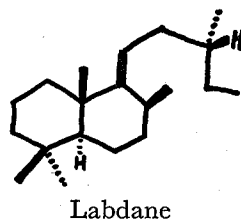
[REF] 1

\* 藤田栄一, 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.

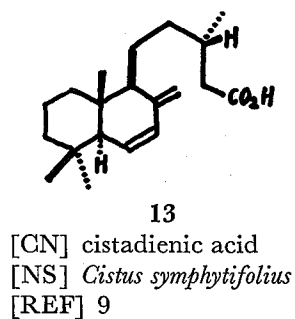
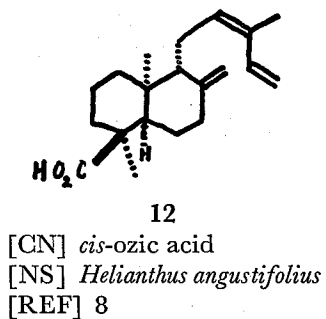
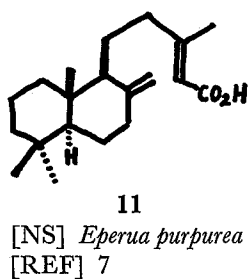
2) Synthesis and Reaction

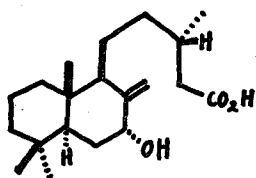


III. LABDANE DERIVATIVES



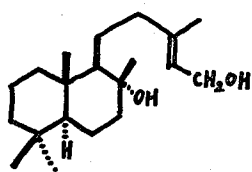
1) Isolation and Structure Determination





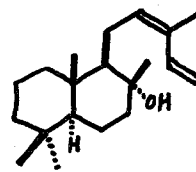
14

[CN] cistenolic acid  
[NS] *Cistus symphytifolius*  
[REF] 9



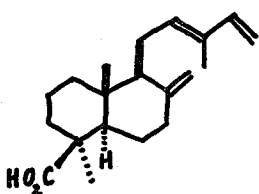
15

[NS] *Cistus symphytifolius*  
[REF] 9



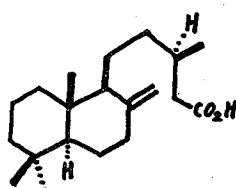
16

[CN] (+)-abienol  
[NS] *Fleischmannia pycnocephaloides*  
[REF] 10



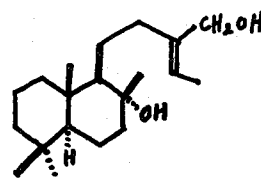
17

[NS] *Fleischmannia deborabellae*  
[REF] 10



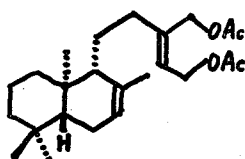
18

[NS] *Eperua purpurea*  
[REF] 11



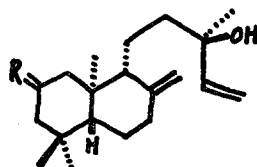
19

[NS] *Picea ajanensis*  
[REF] 12

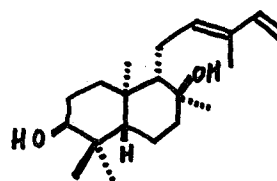


20

[NS] *Baccharis* species  
[REF] 13

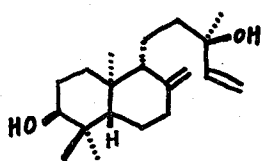


21 R =  $\alpha$ -OH, H  
22 R =  $\beta$ -OH, H  
23 R = 0  
24 R =  $\alpha$ -OCO(CH<sub>2</sub>)<sub>2</sub>CO<sub>2</sub>H, H  
[NS] *Baccharis oxydonta*  
[REF] 14



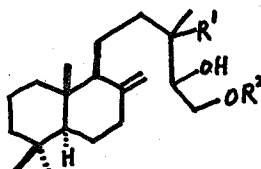
25

[NS] *Grazielia* species  
[REF] 15

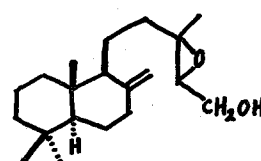


26

[NS] *Croton sublyratus*  
[REF] 16

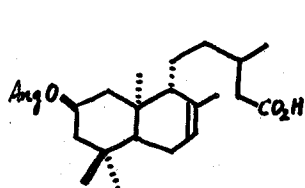


27 R<sup>1</sup> = R<sup>2</sup> = H  
28 R<sup>1</sup> = OH, R<sup>2</sup> = H  
29 R<sup>1</sup> = OH, R<sup>2</sup> = Ac  
[NS] *Hemizonia lutescens*  
[REF] 17



30

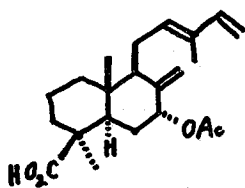
[NS] *Hemizonia lutescens*  
[REF] 17



31

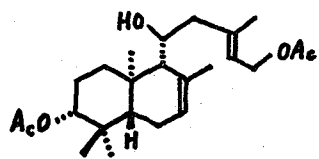
[CN] dihydrodendroidinic acid

[NS] *Pleurocoronis plurisetata*  
[REF] 18



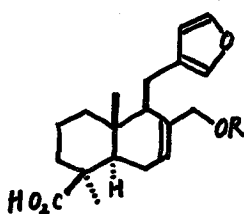
32

[NS] *Chromolaena collina*  
[REF] 19



33

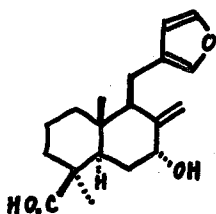
[NS] *Lasiolaena santosii*  
[REF] 20



34 R=H

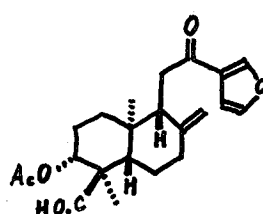
35 R=Ac

[NS] *Gutierrezia dracunculoides*  
[REF] 21



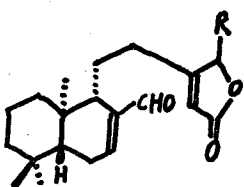
36

[NS] *Gutierrezia dracunculoides*  
[REF] 21



37

[NS] *Dodonaea petiolaris*  
[REF] 22

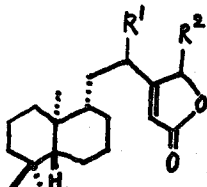


38 R=β-OH

[CN] acritolongifolide A

39 R=α-OH

[CN] acritolongifolide B  
[NS] *Acritopappus longifolius*  
[REF] 23



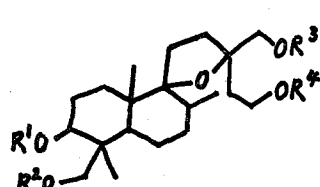
40a R<sup>1</sup>=H, R<sup>2</sup>=α-OH

40b R<sup>1</sup>=H, R<sup>2</sup>=β-OH

41a R<sup>1</sup>=OH, R<sup>2</sup>=α-OH

41b R<sup>1</sup>=OH, R<sup>2</sup>=β-OH

[NS] *Ageratum fastigiatum*  
[REF] 24

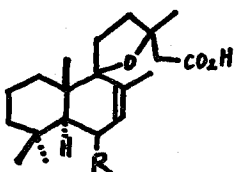


42 R<sup>1</sup>=R<sup>3</sup>=Ac, R<sup>2</sup>=R<sup>4</sup>=H

43 R<sup>1</sup>=R<sup>4</sup>=Ac, R<sup>2</sup>=R<sup>3</sup>=H

44 R<sup>1</sup>=R<sup>3</sup>=H, R<sup>2</sup>=R<sup>4</sup>=Ac

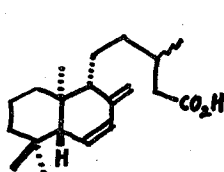
[NS] *Lagochilus inebrians*  
[REF] 25



45 R=α-OH

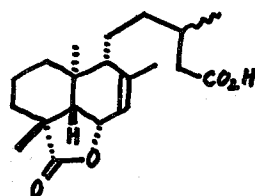
46 R=β-OH

[NS] *Grindelia humilis*  
[REF] 26



47

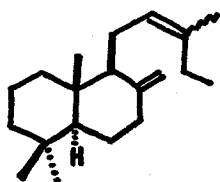
[NS] *Hartwrightia floridana*  
[REF] 27



48

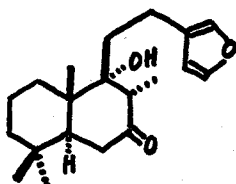
[NS] *Hartwrightia floridana*  
[REF] 27

2) Synthesis and Reactions



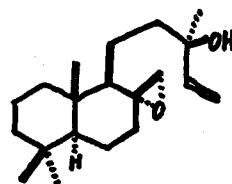
49

[REF] 28  
[NC] photosensitized  
oxygenation of 49  
and related compds.



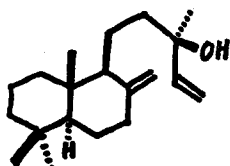
50

[CN] hispanolone  
[REF] 29, 30, 31  
[NC] chemical trans-  
formation of 50



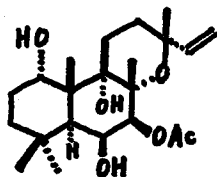
51

[REF] 32  
[NC] acid-catalysed  
rearrangement



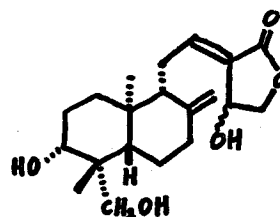
52

[CN] manool  
[REF] 33  
[NC] conversion to  
isoagatholactone



53

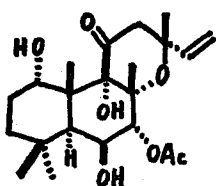
[CN] forskolin  
[NS] *Coleus forskohlii*  
[REF] 34  
[NC] reactions



54

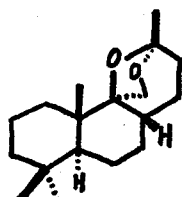
[CN] andrographolide  
[REF] 35  
[NC] reaction with  
NaHSO<sub>3</sub>

3) Miscellaneous Section



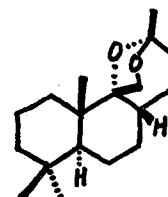
55

[CN] coleonol  
[NS] *Coleus forskohlii*  
[REF] 36  
[NC] pharmacological  
studies



56

[REF] 37  
[NC] •synthesis from  
manool  
•odour studies



57

[REF] 37  
[NC] •synthesis from  
manool  
•odour studies

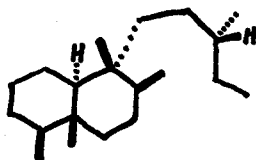
Additional references

[REF] 38 - 41  
[NC] <sup>13</sup>C NMR studies

[REF] 42  
[NC] studies on optical  
activity

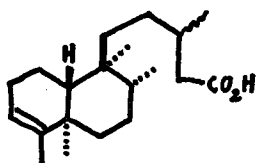
[REF] 43  
[NC] separation by HPLC

IV. CLERODANE DERIVATIVES



Clerodane

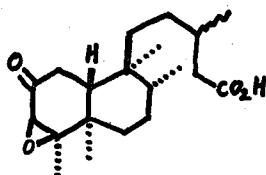
1) Isolation and Structure Determination



58

[NS] *Hartwrightia floridana*

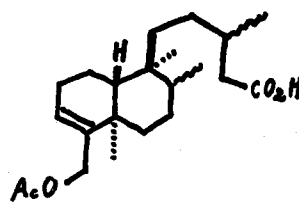
[REF] 27



59

[NS] *Hartwrightia floridana*

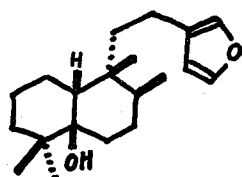
[REF] 27



60

[NS] *Liatris scariosa*

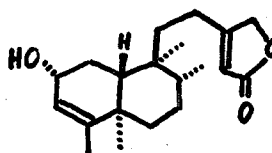
[REF] 44



61

[NS] *Dysidea ambia*

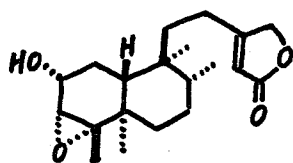
[REF] 45



62

[NS] *Symphiopappus compressus*

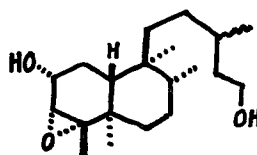
[REF] 46



63

[NS] *Symphiopappus compressus*

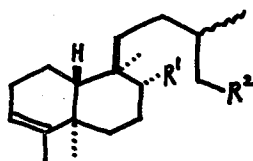
[REF] 46



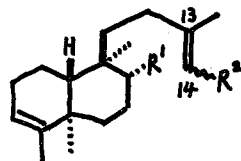
64

[NS] *Symphiopappus compressus*

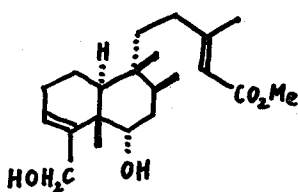
[REF] 46



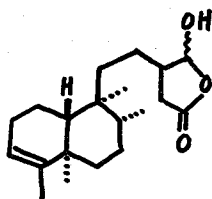
- 65  $R^1 = \text{CH}_2\text{OH}$ ,  $R^2 = \text{CHO}$   
 66  $R^1 = \text{CH}_2\text{OH}$ ,  $R^2 = \text{CH}_2\text{OH}$   
 67  $R^1 = \text{CH}_2\text{OH}$ ,  
 $R^2 = \text{CH}_2\text{OCO}(\text{CH}_2)_{18}\text{Me}$   
 68  $R^1 = \text{CH}_2\text{OH}$ ,  
 $R^2 = \text{CH}_2\text{OCO}(\text{CH}_2)_{20}\text{Me}$   
 69  $R^1 = \text{CHO}$ ,  $R^2 = \text{CH}_2\text{OH}$   
 [NS] *Symphypappus reticulatus*  
 [REF] 46



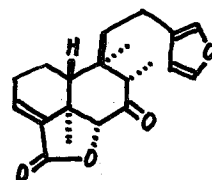
- 70  $R^1 = \text{CH}_2\text{OH}$ ,  $R^2 = \text{CHO}$   
 (13, 14 E and Z)  
 71  $R^1 = \text{CO}_2\text{H}$ ,  $R^2 = \text{CHO}$   
 (13, 14 E and Z)  
 72  $R^1 = R^2 = \text{CH}_2\text{OH}$  (13, 14 E)  
 73  $R^1 = \text{CH}_2\text{OH}$ ,  
 $R^2 = \text{CH}_2\text{OCO}(\text{CH}_2)_{18}\text{Me}$  (13, 14 E)  
 74  $R^1 = \text{CH}_2\text{OH}$ ,  
 $R^2 = \text{CH}_2\text{OCO}(\text{CH}_2)_{20}\text{Me}$  (13, 14 E)  
 [NS] *Symphypappus reticulatus*  
 [REF] 46



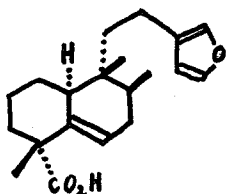
75  
 [NS] *Pityrodia lepidota*  
 [REF] 47



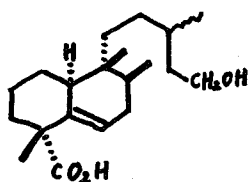
76  
 [NS] *Balianthus viscidus*  
 [REF] 48



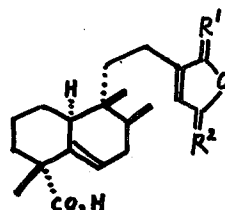
77  
 [NS] *Pulicaria gnaphalodes*  
 [REF] 49



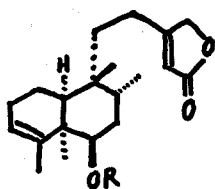
78  
 [CN] koanophyllic acid A  
 [NS] *Koanophyllon*  
*conglobatum*  
 [REF] 50



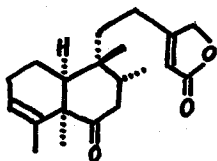
79  
 [CN] koanophyllic acid B  
 [NS] *Koanophyllon*  
*conglobatum*  
 [REF] 50



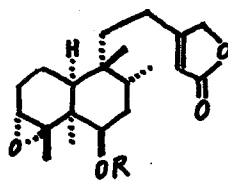
80  $R^1 = \text{O}$ ,  $R^2 = \text{H}_2$   
 [CN] koanophyllic acid C  
 81  $R^1 = \text{H}_2$ ,  $R^2 = \text{O}$   
 [CN] koanophyllic acid D  
 [NS] *Koanophyllon*  
*conglobatum*  
 [REF] 50



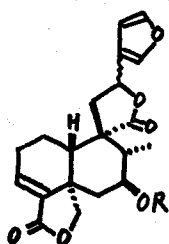
- 82 R=angeloyl  
[CN] solidagolactone II  
83 R=tigloyl  
[CN] solidagolactone III  
84 R=H  
[CN] solidagolactone IV  
85 R=Ac  
[CN] solidagolactone VIII  
[REF] 51  
[NC] structure revision



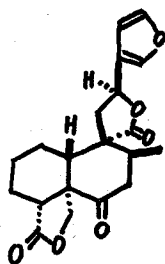
- 86  
[CN] solidagolactone V  
[REF] 51  
[NC] structure revision



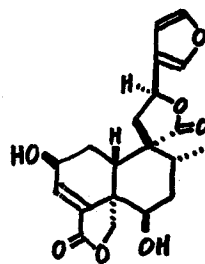
- 87 R=Ac  
[NC] solidagolactone VI  
88 R=angeloyl  
[CN] solidagolactone VII  
[REF] 51  
[NC] structure revision



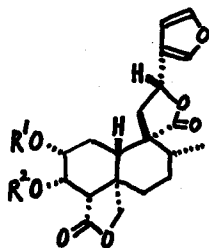
- 89 R=angeloyl  
[NS] *Baccharis subdentata*  
[REF] 14



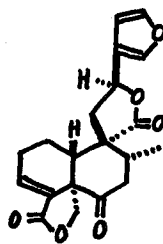
- 90  
[NS] *Teucrium scordium*  
[REF] 52



- 91  
[CN] teugin  
[NS] *Teucrium fragile*  
[REF] 53

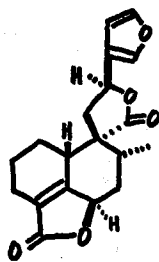


- 92 R<sup>1</sup>=angeloyl, R<sup>2</sup>=senecioyl  
93 R<sup>1</sup>=senecioyl, R<sup>2</sup>=angeloyl  
94 R<sup>1</sup>=angeloyl, R<sup>2</sup>=2-methylbutyl  
[NS] *Baccharis* species  
[REF] 13



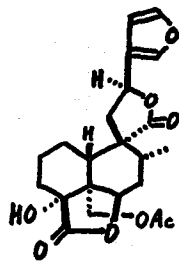
- 95  
[CN] teuscordinon  
[NS] *Teucrium scordium*  
[REF] 54





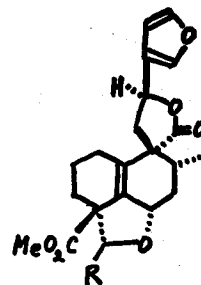
96

[CN] teuffin  
[NS] *Teucrium viscidum*  
var. *Miguelianum*  
[REF] 55



97

[NS] *Teucrium* species  
[REF] 56



98 R =  $\beta$ -OH

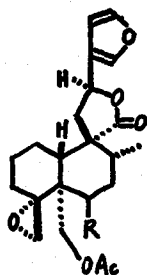
[CN] mallotucin C

99 R =  $\alpha$ -OH

[CN] mallotucin D

[NS] *Mallotus repandus*

[REF] 57



100 R =  $\beta$ -OH

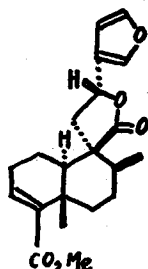
[CN] teucjaponin A

101 R =  $\alpha$ -OH

[CN] teucjaponin B

[NS] *Truvtium japonicum*

[REF] 58

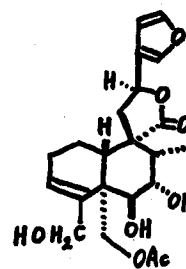


102

[CN] sonderianin

[NS] *Croton sonderianus*

[REF] 59

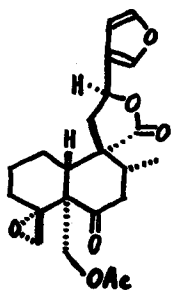


103

[CN] lolin

[NS] *Teucrium capitatum*

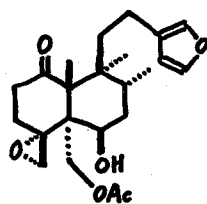
[REF] 60



104

[NS] *Teucrium gnaphalodes*

[REF] 61

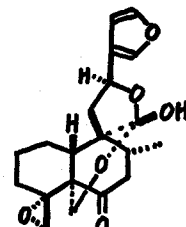


105

[CN] isofruticolone

[NS] *Teucrium gnaphalodes*

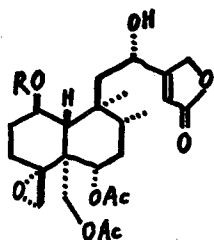
[REF] 61



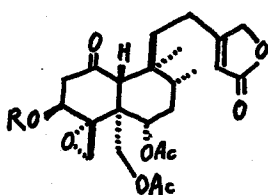
106

[NS] *Teucrium gnaphalodes*

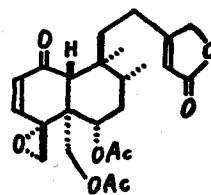
[REF] 61



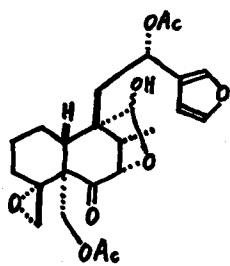
107  $R = \text{COC}(\text{Me}) = \text{CHMe}$   
 [CN] ajugamarin  
 [NS] *Ajuga nipponensis*  
 [REF] 62



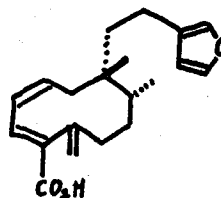
108  $R = \text{COCH}(\text{Me})\text{Et}$   
 [CN] ajugareptansone A  
 [NS] *Ajuga reptans*  
 [REF] 63



109  
 [CN] ajugareptansone B  
 [NS] *Ajuga reptans*  
 [REF] 63

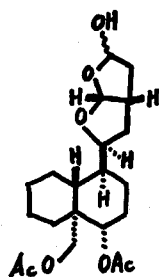


110  
 [CN] auropolin  
 [NS] *Teacrium polium*  
 snbsp. *aureum*  
 [REF] 64

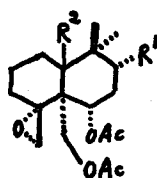


111  
 [CN] conyzic acid  
 [NS] *Conyza strict*  
 [REF] 65

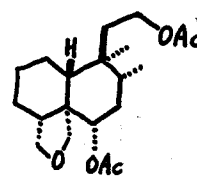
## 2) synthesis and Reactions



112  
 [REF] 66  
 [NC] •synthesis  
 •insect  
 antifeedant



113  $R^1 = \text{H}, R^2 = \beta\text{-H}$   
 114  $R^1 = \text{H}, R^2 = \alpha\text{-H}$   
 115  $R^1 = \text{Me}, R^2 = \beta\text{-H}$   
 [REF] 67 - 69  
 [NC] •synthesis  
 •insect  
 antifeedant



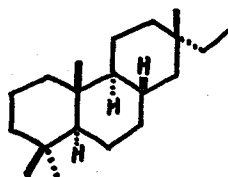
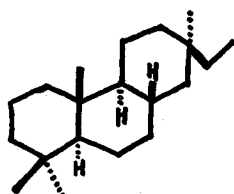
116  
 [REF] 70  
 [NC] synthesis

### 3) Miscellaneous Section

[REF] 71

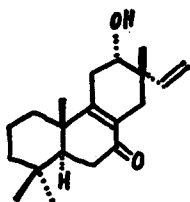
[NC] studies on insect antifeeding activity

## V. PIMARANE AND ISOPIMARANE DERIVATIVES



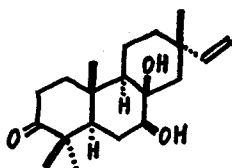
Pimarane and Isopimarane

### 1) Isolation and Structure Determination



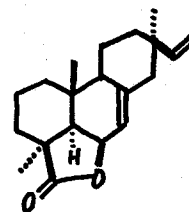
117

[NS] *Vellozia* species  
[REF] 72



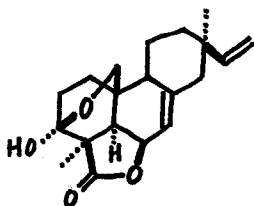
118

[NS] *Bromelia*  
*pinguin*  
[REF] 73



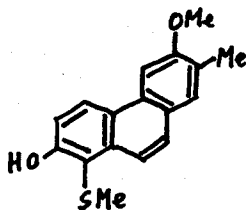
119

[CN] momilactone A  
[NS] rice  
[REF] 74  
[NC] phytoalexin



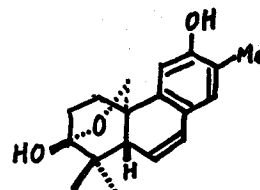
120

[CN] momilactone B  
[NS] rice  
[REF] 74  
[NC] phytoalexin



121

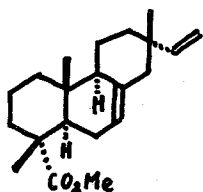
[CN] micrandrol C  
[NS] *Micrandropsis*  
*scleroxylon*  
[REF] 75



122

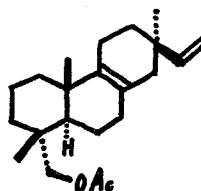
[CN] micrandrol D  
[NS] *Micrandropsis*  
*scleroxylon*  
[REF] 75

2) Synthesis and Reaction



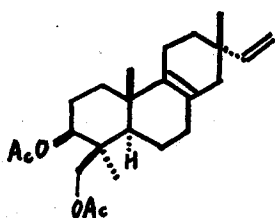
123

[REF] 76  
[NC] superacid catalyzed cyclization



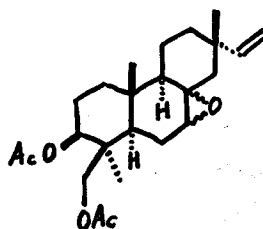
124

[REF] 77  
[NC] photo-oxygenation



125

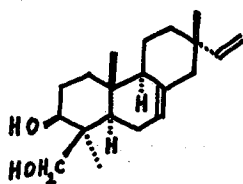
[REF] 77  
[NC] photo-oxygenation



126

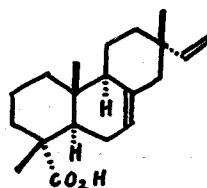
[REF] 78  
[NC] chemical transformation

3) Miscellaneous Section



127

[CN] virescenol B  
[REF] 79  
[NC] biosynthesis



128

[CN] isopimaric acid  
[REF] 80  
[NC] biotransformation

Additional references

[REF] 81 - 83

[NC] <sup>13</sup>C NMR studies

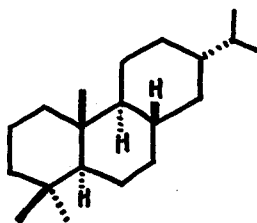
[REF] 84

[NC] studies on distribution of podolactone-type plant growth inhibitors

[REF] 85

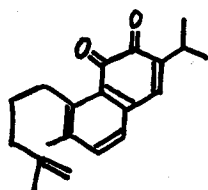
[NC] studies on conformations for pimarane derivatives

VI. ABIETANE DERIVATIVES



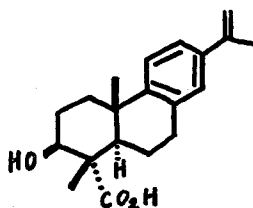
Abietane

1) Isolation and Structure Determination



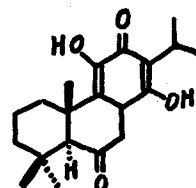
129

[CN] aethiopinone  
[NS] *Salvia aethiopsis*  
[REF] 86



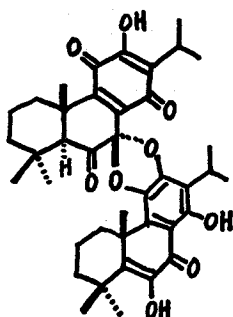
130

[NS] *Salvia tomentosa*  
[REF] 87

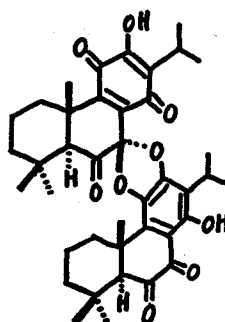


131

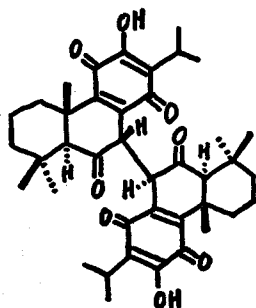
[NS] *Plectranthus grandidentatus*  
[REF] 88



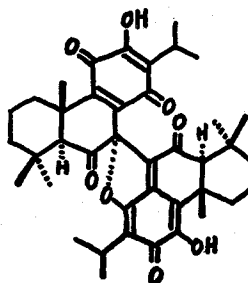
132 [CN] grandidone A  
133 [CN] 7-epigrandidone A  
[NS] *Plectranthus grandidentatus*  
[REF] 88



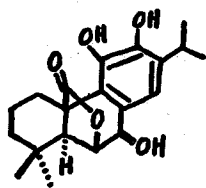
134 [CN] grandidone B  
135 [CN] 7-epigrandidone B  
[NS] *Plectranthus grandidentatus*  
[REF] 88



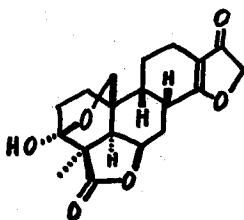
136  
[CN] grandidone C  
[NS] *Plectranthus grandidentatus*  
[REF] 88



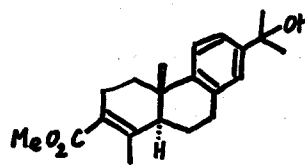
137 [CN] grandidone D  
138 [CN] 7-epigrandidone D  
[NS] *Plectranthus grandidentatus*  
[REF] 88



139  
[CN] rosmanol  
[NS] *Rosmarinus officinalis*  
[REF] 89  
[NC] antioxidant

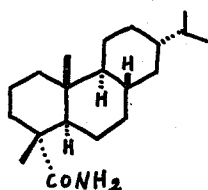


140  
[CN] humiliathenolide C  
[NS] *Humirianthera rupestris*  
[REF] 1

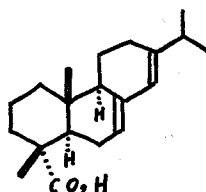


141  
[NS] *Tripterygium wilfordii*  
[REF] 90  
[NC] isolation of cytotoxic diterpenes. (tripdio-loide, triptolide, etc.)

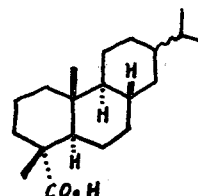
## 2) Synthesis and Reaction



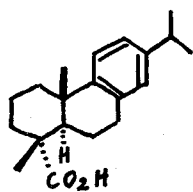
142  
[REF] 91  
[NC] lactonization  
by photolysis  
( $\text{Pb}(\text{OAc})_4/\text{I}_2$ )



143  
[CN] abietic acid  
[REF] 92  
[NC] air oxidation

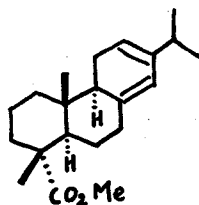


144  
[REF] 93  
[NC] carbonylation by  
conc.  $\text{H}_2\text{SO}_4$



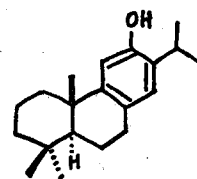
145

- [CN] dehydroabietic acid  
 [REF] 94  
 [NC] oxidation by  
 Co(acac)<sub>2</sub>  
 [REF] 95  
 [NC] Synthesis of (+)-  
 fragrolide and (+)-  
 bemadienolide



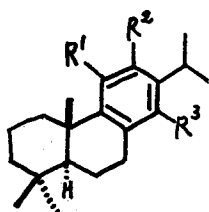
146

- [CN] methyl levopimarate  
 [REF] 96  
 [NC] addition of  
 ClSO<sub>2</sub>NCO



147

- [CN] ferruginol  
 [REF] 97  
 [NC] synthesis from 145  
 [REF] 98  
 [NC] ozonolysis

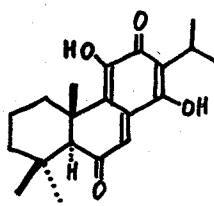


148 R<sup>1</sup>=R<sup>2</sup>=OH, R<sup>3</sup>=H

149 R<sup>1</sup>=R<sup>2</sup>=H, R<sup>3</sup>=OH

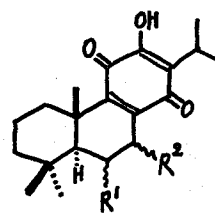
[REF] 98

- [NC] Chemical conversion  
 to isodrimenin, vali-  
 divioloide, winterin,  
 and confertifolin



150

- [CN] 14-hydroxytaxodione  
 [REF] 99  
 [NC] partial synthesis and  
 reactions of 150

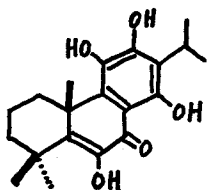


151 R<sup>1</sup>=R<sup>2</sup>=α-OH

152 R<sup>1</sup>=α-OH,  
 R<sup>2</sup>=β-OH

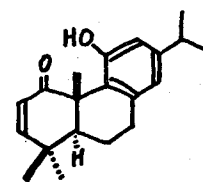
153 R<sup>1</sup>=R<sup>2</sup>=β-OH  
 [REF] 100

- [NC] partial synthesis



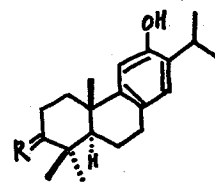
154

- [CN] coleoneU  
 [REF] 101 and 102  
 [NC] synthesis



155

- [REF] 103  
 [NC] synthesis



156 R=α-H, β-OH

[CN] hinokiol

157 R=O

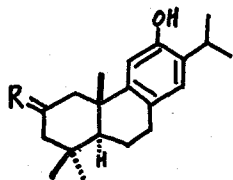
[CN] hinokione

[REF] 104

[NC] synthesis from  
 abietatrienol

[REF] 105

- [NC] total synthesis



158 R =  $\alpha$ -OH,  $\beta$ -H

[CN] salviol

159 R = O

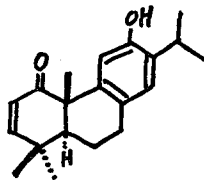
[CN] 2-oxoferruginol

[REF] 104

[NC] synthesis from abietatrienol

[REF] 105

[NC] total synthesis



160

[CN] shonanol

[REF] 106

[NC] synthesis and revised structure

131

[REF] 107

[NC] partial synthesis

(131-138)

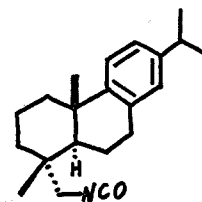
from 131

### 3) Miscellaneous Section

145

[REF] 108

[NC] biodegradation of dehydroabietic acid (145) with *Mortierella isabellina*

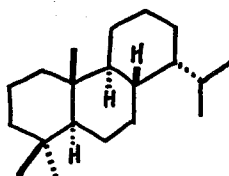


161

[REF] 109

[NC] use in the synthesis of (*S*)-5-hydroxy-6-*trans*-8, 11, 14-*cis*-eicosatetraenoic acid

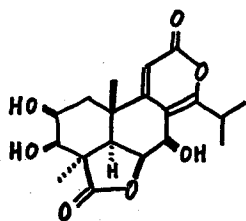
## VII. TOTARANE DERIVATIVES



Totarane

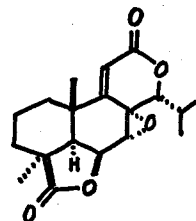


1) Isolation and Structure Determination



162

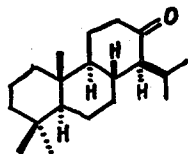
[CN] urbalactone  
[NS] *Podocarpus urbanii*  
[REF] 110



163

[CN] 2,3-dihydropodolide  
[NS] *Podocarpus urbanii*  
[REF] 110

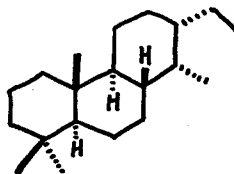
2) Synthesis and Reaction



164

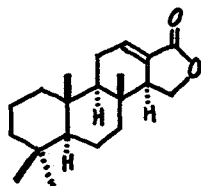
[REF] 111  
[NC] Synthesis *via* cyclobutane ring-opening  
of allene-enone photoadduct

VIII. CASSANE DERIVATIVES



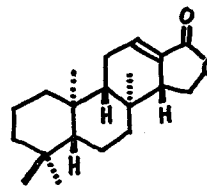
Cassane

1) Synthesis and Reaction



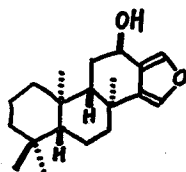
165

[CN] (+)-isoagatholactone  
[REF] 33  
[NC] synthesis from (+)-manool



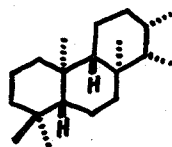
166

[CN] *ent*-isoagatholactone  
[REF] 112  
[NC] synthesis from methyl isocopalate



167

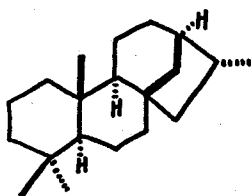
[CN] *ent*-13 (16),14-spongiadien-12 $\alpha$ -ol  
 [REF] 112  
 [NC] synthesis from methyl isocopalate



168

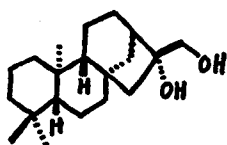
[CN] isocopalane  
 [REF] 112  
 [NC] synthesis

### IX. KAURANE DERIVATIVES



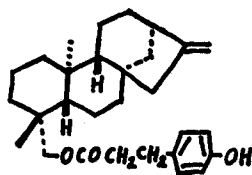
Kaurane

#### 1) Isolation and Structure Determination



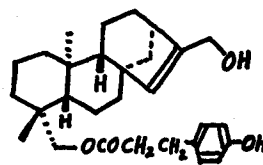
169

[NS] *Croton sublyratus*  
 [REF] 16  
 [NS] *Aristolachia elegans*  
 [REF] 113



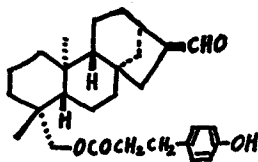
170

[NS] *Baccharis quitensis*  
 [REF] 14



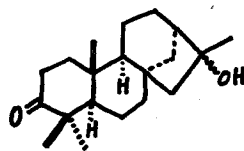
171

[NS] *Baccharis quitensis*  
 [REF] 14



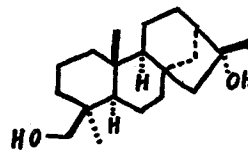
172

[NS] *Baccharis quitensis*  
 [REF] 14



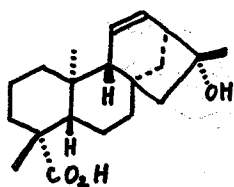
173

[NS] *Bromelia pinguin*  
 [REF] 73

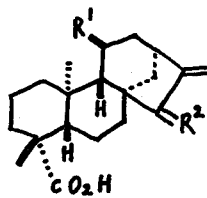


174

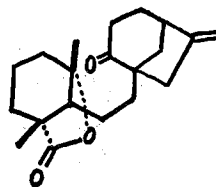
[NS] *Bromelia pinguin*  
 [REF] 73



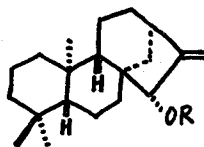
175  
[NS] *Helianthus angustifolius*  
[REF] 8



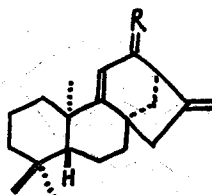
176 R<sup>1</sup>=H,  
R<sup>2</sup>= $\alpha$ -OTigl  
177 R<sup>1</sup>=OH, R<sup>2</sup>=O  
178 R<sup>1</sup>=OH,  
R<sup>2</sup>= $\beta$ -OH, H  
[NS] *Grazielia* species  
[REF] 15



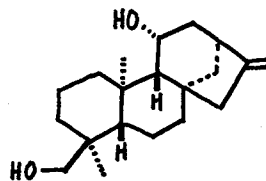
179  
[NS] *Ageratum fastigiatum*  
[REF] 24



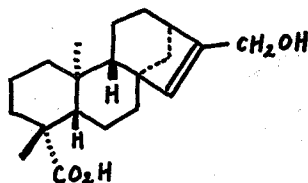
R=methylacryloyl  
180  
[NS] *Ichthyothere* species  
[REF] 114



181 R= $\alpha$ -H,  $\beta$ -OH  
182 R=O  
[NS] *Vellozia caput-ardeae*  
[REF] 115

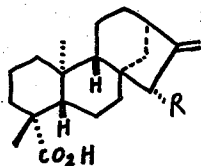


183  
[NS] *Sideritis arborescens*  
[REF] 116



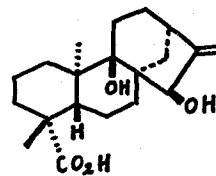
184

[NS] *Helianthus grosseserratus*  
[REF] 117

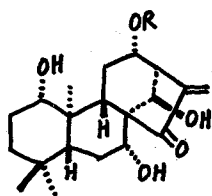


185 R=OH  
[CN] grandifloric acid  
[NS] *Helianthus grosseserratus*  
[REF] 117  
186 R=H  
[NS] *Aristolochia triangularis*  
[REF] 118  
[NS] *Wedelia buphthalmiflora*  
[REF] 119

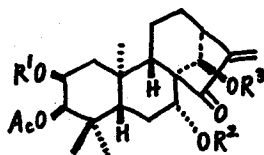
187 R=H; 4<sup>9(11)</sup>  
[CN] grandiflorenic acid  
188 R=MeCH=CMeCOO-(Z)  
189 R=MeCH=CMeCOO-(E)  
190 R=Me<sub>2</sub>CHCH<sub>2</sub>COO-  
191 R=Me<sub>2</sub>CHCOO-  
[NS] *Wedelia buphthalmiflora*  
[REF] 119



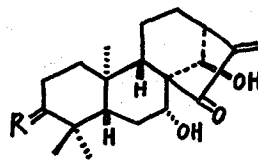
192  
[CN] pterokaurene L<sub>2</sub>  
[NS] *Pteris longipes*  
[REF] 120



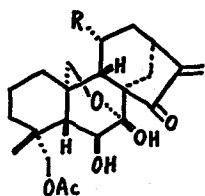
- 193 R=H  
[CN] excisanin A  
194 R=Ac  
[CN] excisanin B  
[NS] *Rabdosia excisa*  
[REF] 121



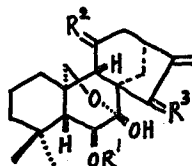
- 195 R<sup>1</sup>=R<sup>2</sup>=R<sup>3</sup>=H  
[CN] leukamenin A  
196 R<sup>1</sup>=Ac, R<sup>2</sup>=R<sup>3</sup>=H  
[CN] leukamenin B  
197 R<sup>1</sup>=R<sup>3</sup>=Ac, R<sup>2</sup>=H  
[CN] leukamenin C  
198 R<sup>1</sup>=R<sup>2</sup>=Ac, R<sup>3</sup>=H  
[CN] leukamenin D  
[NS] *Rabdosia umbrosa*  
*var. leucantha f. kameba*  
[REF] 122



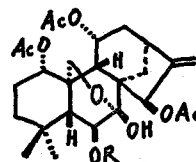
- 199 R= $\alpha$ -H,  $\beta$ -OAc  
[CN] leukamenin E  
200 R=O  
[CN] leukamenin F  
[NS] *Rabdosia umbrosa*  
*var. leucantha f. kameba*  
[REF] 122



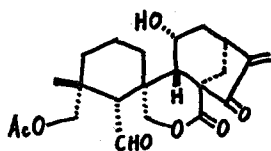
- 201 R=H  
[CN] longikaurin C  
202 R=OH  
[CN] longikaurin D  
203 R=OAc  
[CN] longikaurin F  
[NS] *Rabdosia longituba*  
[REF] 123  
[NC] antibacterial  
diterpenoids



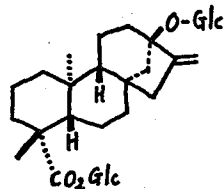
- 204 R<sup>1</sup>=H, R<sup>2</sup>= $\alpha$ -H,  
 $\beta$ -OAc, R<sup>3</sup>=O  
[CN] longikaurin E  
[NS] *Rabdosia longituba*  
[REF] 123  
205 R<sup>1</sup>=OAc, R<sup>2</sup>=O,  
R<sup>3</sup>= $\alpha$ -H,  $\beta$ -OH  
[CN] rabdosianin C  
[NS] *Rabdosia shikokiana*  
[REF] 124



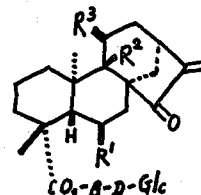
- 206 R=H  
[CN] rabdosianin A  
207 R=Ac  
[CN] rabdosianin B  
[NS] *Rabdosia shikokiana*  
[REF] 124



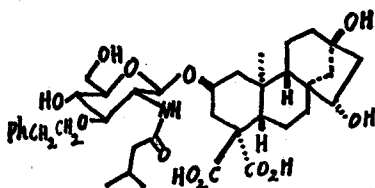
- 208  
[CN] trichorabdal B  
[NS] *Rabdosia trichocarpa*  
[REF] 125  
[NC] X-ray crystal structure  
and transformation  
into novel skeleton



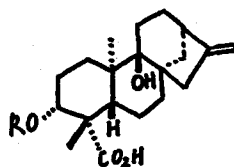
- 209  
[CN] rubusoside  
[NS] *Rubus chingii*  
[REF] 126  
[NC] sweet principle



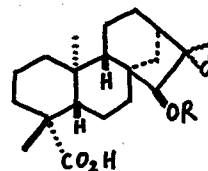
- 210 R<sup>1</sup>=R<sup>3</sup>=H, R<sup>2</sup>=OH  
211 R<sup>1</sup>=R<sup>3</sup>=OH, R<sup>2</sup>=H  
212 R<sup>1</sup>=R<sup>2</sup>=OH, R<sup>3</sup>=H  
[NS] *Pteris livida*  
[REF] 127



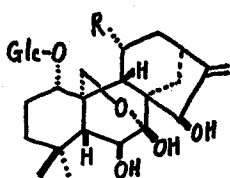
213  
[CN] wedeloside  
[NS] *Wedelia asperima*  
[REF] 128 and 129  
[NC] toxicity and potential  
antitumor activity



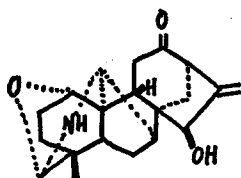
214 R=Ang  
215 R=Cinn  
[NS] *Wedelia trilobata*  
[REF] 130



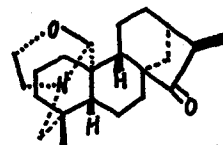
216 R=Ang  
217 R=Tigl  
[NS] *Aspilia parvifolia*  
[REF] 130



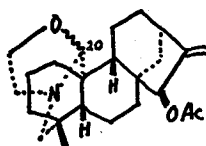
218 R=OH  
[CN] shikokiaside A  
219 R=H  
[CN] shikokiaside B  
[NS] *Rabdosia shikokiana*  
*var. shikokiana*  
[REF] 131



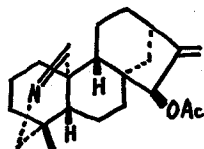
220  
[CN] norsongoramine  
[NS] *Delphinium tamarae*  
[REF] 132



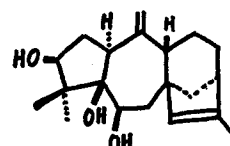
221  
[CN] cuachichicine  
[REF] 133  
[NC] revised structure and  
<sup>13</sup>C-NMR and X-ray  
crystallography



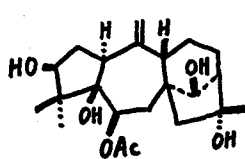
222  
[CN] ovatine  
[NS] *Garrya ovata* var.  
*lindheimeri*  
[REF] 133  
[NC] mixture of C(20)  
epimer



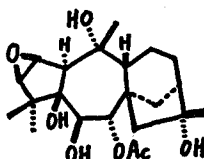
223  
[CN] lindheimerine  
[NS] *Garrya ovata* var.  
*lindheimeri*  
[REF] 133



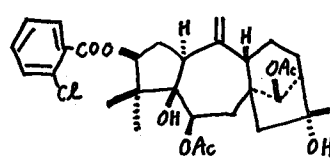
224  
[CN] grayanotoxin XIX  
[NS] *Leucothoe grayana*  
[REF] 134



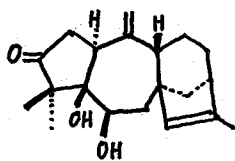
225  
[CN] grayanotoxin XVI  
[NS] *Leucothoe grayana*  
[REF] 135



226  
[CN] lyoniatoxin  
[NS] *Lyonia ovalifolia*  
*var. elliptica*  
[REF] 136  
[NC] revised structure

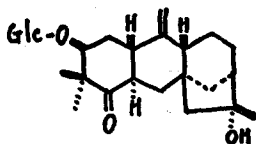


227  
[REF] 137  
[NC] X-ray analysis and  
boat conformation  
of C-ring



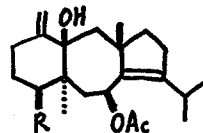
228

[CN] grayanotoxin XVIII  
[NS] *Leucothoe grayana*  
[REF] 138  
[NC] X-ray analysis



229

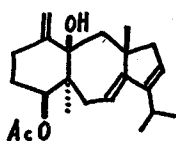
[CN] pieroside B  
[NS] *Pieris japonica*  
[REF] 139  
[NC] first example of a  
leucothane glycoside



230 R=H

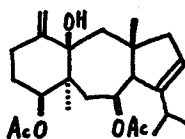
231 R=OH

[NS] *Dictyota divaricata*  
[REF] 140  
[NC] X-ray analysis



232

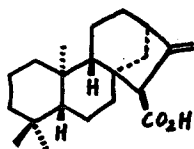
[NS] *Dictyota divaricata*  
[REF] 140  
[NC] X-ray analysis



233

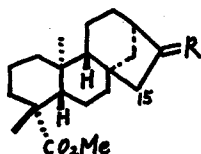
[NS] *Dictyota divaricata*  
[REF] 140  
[NC] X-ray analysis

## 2) Synthesis and Reaction



234

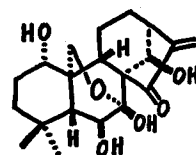
[REF] 141  
[NC] carboxylation of  
kaurene



235 R=F<sub>2</sub>

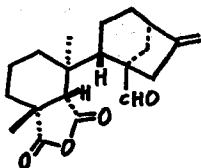
236 R=CF<sub>3</sub> (15-en)

[REF] 142  
[NC] derived from  
xylopic acid



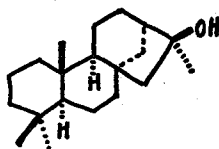
237

[CN] oridonin  
[REF] 143  
[NC] selective acetylation



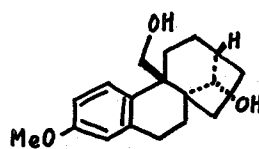
238

[CN] fujenal  
[REF] 144 and 145  
[NC] preparation of some  
7-norgibberellanes  
from 238



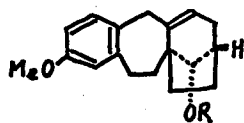
239

[CN] phyllocladan-16β-ol  
[REF] 146  
[NC] synthesis from  
phyllocladene



240

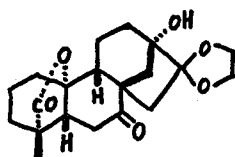
[REF] 147  
[NC] synthetic approach  
to grayanotoxins



241

[REF] 148

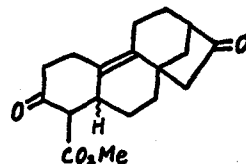
[NC] synthetic approach to grayanotoxins



242

[REF] 149

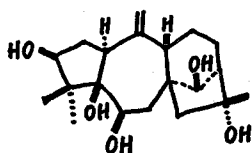
[NC] synthetic approach to gibberellins



243

[REF] 150

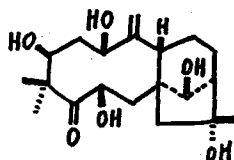
[NC] synthetic approach to gibberellins



244

[REF] 151

[NC] ring opening with  $Pb(OAc)_4$

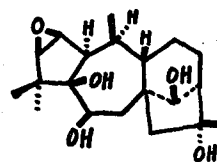


245

[CN] grayanol B

[REF] 152

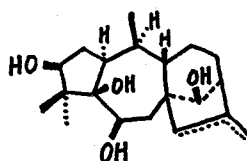
[NC] synthesis from 243



246

[REF] 153

[NC] conversion of 244 to 246 and 247

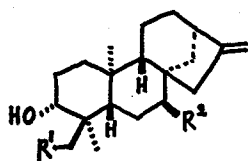


247

[REF] 153

[NC] conversion of 244 to 246 and 247

### 3) Miscellaneous Section



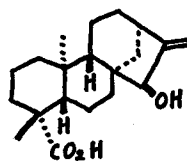
248  $R^1=R^2=H$

249  $R^1=OH, R^2=H$

250  $R^1=R^2=OH$

[REF] 154

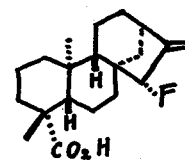
[NC] microbial transformation of 248-250



251

[REF] 155

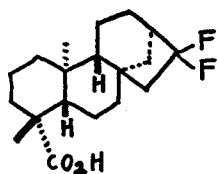
[NC] microbial production of plant gibberellins from 251 in *G. Fujikuroi*



252

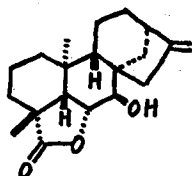
[REF] 156

[NC] microbial production of fluoro gibberellins from 252



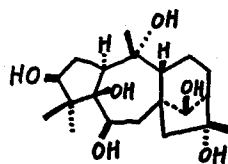
253

[REF] 157  
[NC] microbial transformation



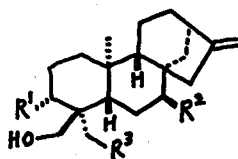
254

[REF] 158  
[NC] kaurenolide biosynthesis



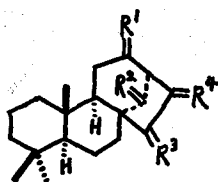
255

[CN] grayanotoxin III  
[REF] 159  
[NC] biosynthesis of grayanotoxin



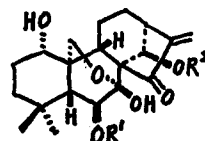
256

[REF] 160  
[NC] <sup>13</sup>C NMR studies



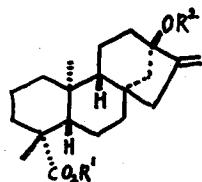
257

[REF] 161  
[NC] <sup>13</sup>C NMR studies



258

[REF] 162  
[NC] antitumor activity of acylated oridonin



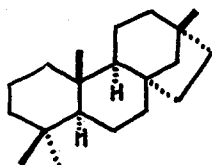
259

[CN] rebandioside-A, D, E  
[REF] 163  
[NC] structure-sweetness relationship and synthesis

Additional references

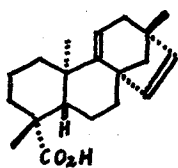
[REF] 164  
[NC] sensory evaluation of stevioside analogues

## X. BEYERANE DERIVATIVES



Beyerane

### 1) Isolation and Structure Determination

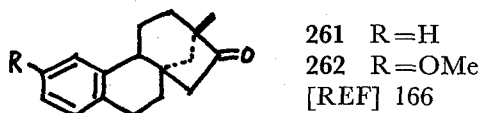


260

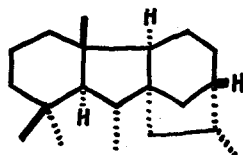
[CN] dehydrostachemic acid  
[NS] *Viguiera* species  
[REF] 165



2) Synthesis and Reaction

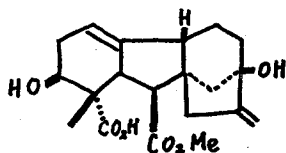


XI. GIBBERELLANE DERIVATIVES



Gibberellane

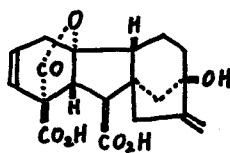
1) Isolation and Structure Determination



263

[REF] 167

[NC] X-ray analysis



264

[CN] gibberellin A<sub>59</sub>

[NS] *Canavalia gladiata*

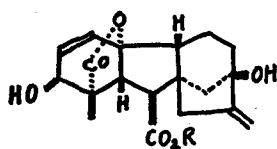
[REF] 168

Additional reference

[NS] *Chrysanthemum morifolium*

[REF] 169

2) Synthesis and Reaction



265

[CN] gibberellin A<sub>3</sub> (R=H)

[REF] 170

[NC] degradation

[REF] 171 (R=H)

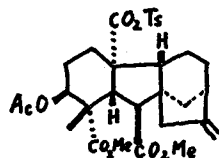
[NC] chemical conversion to GA<sub>5</sub>

[REF] 172 (R=Me)

[NC] reaction with PPh<sub>3</sub>

[REF] 173 (R=Me)

[NC] chemical conversion to GA<sub>9</sub> and GA<sub>20</sub>



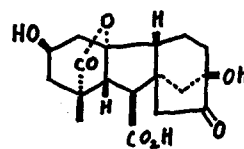
266

[CN] dimethyl ester of gibberellin A<sub>13</sub>

20-toluene-*p*-sulfonyl anhydride

[REF] 174

[NC] methanolysis, reduction with NaBH<sub>4</sub>



267

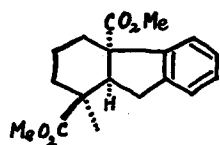
[CN] gibberellin A<sub>29</sub>

[REF] 175

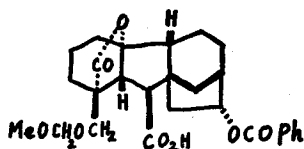
[NC] deuterated and tritiated GA<sub>29</sub>

[REF] 176

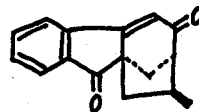
[NC] chemical conversion



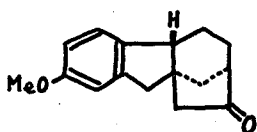
268  
[REF] 177  
[NC] synthesis



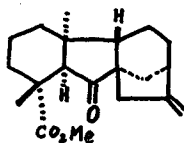
269  
[REF] 178  
[NC] synthesis



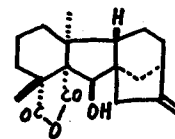
270  
[REF] 179  
[NC] synthesis



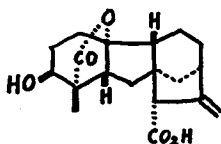
271  
[REF] 180  
[NC] synthesis



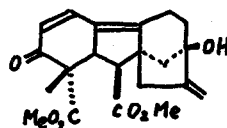
272  
[REF] 144  
[NC] preparation from fujenal



273  
[REF] 144  
[NC] preparation from fujenal



274  
[CN] 7 (6→15βH) *abeo*-gibberellin A<sub>4</sub>  
[REF] 181  
[NC] preparation form GA<sub>4</sub>

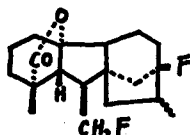


275  
[REF] 182  
[NC] photosensitized dimerization  
[REF] 183  
[NC] photoreduction  
[REF] 184  
[NC] photocycloaddition

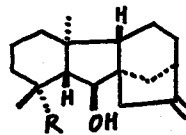
Additional references

- [REF] 185 [REF] 186 [REF] 187 [REF] 188  
[NC] chlorination [NC] glucosylation [NC] methylenation [NC] reduction with K-selectride

3) Miscellaneous Section



276  
[REF] 189  
[NC] feeding experiment

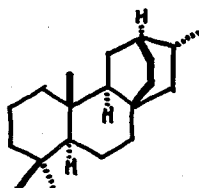


277 R = COOH  
278 R = CH<sub>2</sub>OH  
[REF] 145  
[NC] inhibitors of gibberellin biosynthesis

Additional references

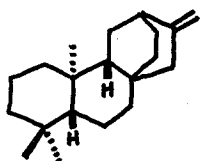
- |   |  |   |
|---|--|---|
| [REF] 109                                       | [REF] 157                                    | [REF] 156                                       |
| [NC] bioassay                                   | [NC] biotransformation of fluorogibberellins | [NC] biotransformation of fluorogibberellins    |
| [REF] 155                                       | [REF] 191                                    | [REF] 192                                       |
| [NC] microbial production of plant gibberellins | [NC] biosynthetic studies                    | [NC] biosynthesis in <i>Phaseolus coccineus</i> |
| [REF] 193                                       | [REF] 194                                    | [REF] 195                                       |
| [NC] effect on the RNA-ase activity             | [NC] gibberellins in callus of crown gall    | [NC] chromatographic separation                 |

XII. ATISANE DERIVATIVES



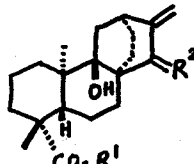
Atisane

1) Isolation and Structure Determination



279

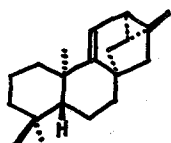
- [CN] atisirene  
[NS] *Thymus capitatus*  
[REF] 196



280

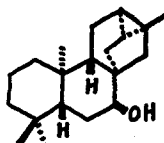
- [CN] pteroatisene P<sub>1</sub> (R<sup>1</sup>=H, R<sup>2</sup>=O)  
pteroatisene P<sub>2</sub> (R<sup>1</sup>=H, R<sup>2</sup>= $\alpha$ -H,  
 $\beta$ -OH)  
pteroatisenoside P<sub>1</sub> (R<sup>1</sup>= $\beta$ -D-glucosyl, R<sup>2</sup>=O)

- [NS] *Pteris purpureorachis*  
[REF] 197, 120  
[NC] X-ray analysis



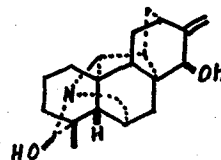
281

- [CN] 9,11-dehydro trachylobanic acid  
[NS] *Viguiera* species  
[REF] 165



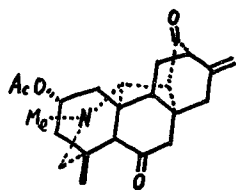
282

- [CN] ciliaric acid  
[NS] *Helianthus grosseserratus*  
[REF] 117



283

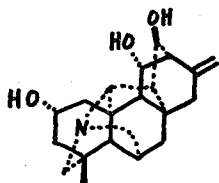
- [CN] talatisene  
[REF] 198  
[NC] X-ray analysis



284

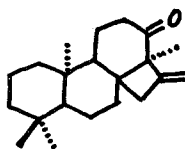
[CN] heterophylloidine  
 [NS] *Aconitum heterophylloides*  
 [REF] 199  
 [NC] X-ray analysis on a derivative

## 2) Synthesis and Reaction



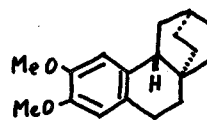
285

[CN] hetisine  
 [REF] 200  
 [NC] rearrangement with acids



286

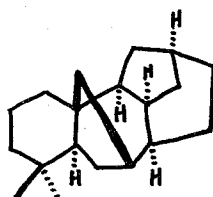
[REF] 201  
 [NC] rearrangement to the  
 atisane skeleton



287

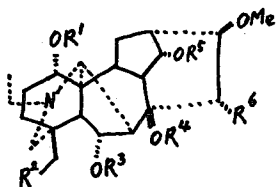
[REF] 202  
 [NC] synthesis from a  
 benzylisoquinoline  
 derivative

## XIII. ACONANE DERIVATIVES



Aconane

### 1) Isolation and Structure Determination



288  $R^1=R^2=R^5=R^6=H$ ,  $R^3=Ac$ ,  
 $R^4=Me$

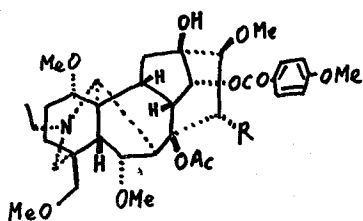
[CN] alkaloid A  
 [NS] *Delphinium bicolor*  
 [REF] 203  
 [NC] X-ray analysis

289  $R^1=R^3=Me$ ,  $R^2=OMe$ ,  $R^4=Ac$ ,  
 $R^5=CO-C_6H_4-OMe$ ,  $R^6=H$

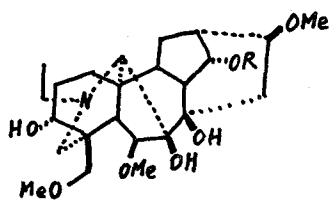
[CN] foresaconitine  
 [NS] *Aconitum forestii*  
 [REF] 204

290  $R^1=R^4=R^5=H$ ,  $R^2=OMe$ ,  
 $R^3=Me$ ,  $R^6=OH$

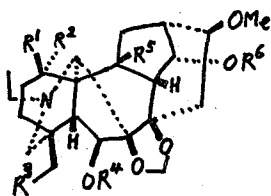
[CN]  $15\alpha$ -hydroxyneoline  
 [NS] *Aconitum* species  
 [REF] 205



- 291 R=H  
 [CN] crassicauline A  
 [NS] *Aconitum crassicaule*  
 [REF] 206  
 292 R=OH  
 [CN] deoxyjsaconitine  
 [NS] *Aconitum subcuneatum*  
 [REF] 207

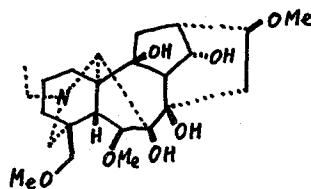


- 294 R=Me (acomonine)  
 295 R=H (iliensine)  
 296 R=-CO- (14-benzoyliliensine)  
 [REF] 209  
 [NC] structure reinvestigation

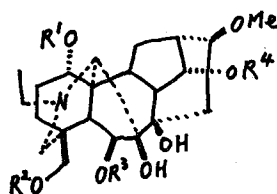


- 302 R<sup>1</sup>=R<sup>3</sup>=OMe, R<sup>2</sup>=R<sup>4</sup>=R<sup>5</sup>=R<sup>6</sup>=H (delcorine)  
 303 R<sup>1</sup>=R<sup>3</sup>=OMe, R<sup>2</sup>=R<sup>4</sup>=R<sup>5</sup>=H, R<sup>6</sup>=Me (delcoridine)  
 [REF] 213

- 304 R<sup>1</sup>=R<sup>3</sup>=H, R<sup>2</sup>=OMe, R<sup>4</sup>=Ac, R<sup>5</sup>=OH, R<sup>6</sup>=CO-CH(CH<sub>3</sub>)CH<sub>2</sub>CH<sub>3</sub> (glaucenine)  
 305 R<sup>1</sup>=R<sup>3</sup>=H, R<sup>2</sup>=OMe, R<sup>4</sup>=Ac, R<sup>5</sup>=OH, R<sup>6</sup>=COCHMe<sub>2</sub> (glaucerine)  
 306 R<sup>1</sup>=R<sup>3</sup>=H, R<sup>2</sup>=OMe, R<sup>4</sup>=Ac, R<sup>5</sup>=OH, R<sup>6</sup>=COPh (glaucephine)  
 [NS] *Delphinium glaucescens*  
 [REF] 212



- 293  
 [CN] delcaroline  
 [NS] *Delphinium carolinianum*  
 [REF] 208



- 297 R<sup>1</sup>=R<sup>3</sup>=R<sup>4</sup>=Me, R<sup>2</sup>=H  
 (lycoctonine)  
 298 R<sup>1</sup>=R<sup>2</sup>=R<sup>3</sup>=Me, R<sup>4</sup>=H  
 (brownine)

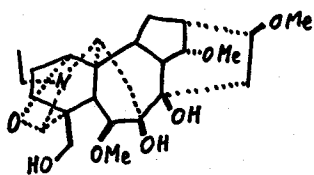
[REF] 210  
 [NC] revision of the stereochemistry at C(1) for 37 alkaloids

- 299 R<sup>1</sup>=R<sup>3</sup>=R<sup>4</sup>=H, R<sup>2</sup>=Me  
 [CN] delphinifoline  
 [NS] *Aconitum delphinifolium*  
 [REF] 211

- [NC] X-ray analysis  
 300 R<sup>1</sup>=R<sup>2</sup>=R<sup>3</sup>=Me,  
 CH<sub>3</sub>  
 R<sup>4</sup>=CO-CH(CH<sub>3</sub>)CH<sub>2</sub>CH<sub>3</sub> (glaucedine)

- 301 R<sup>1</sup>=R<sup>4</sup>=Me, R<sup>2</sup>=,  
 R<sup>3</sup>=H (glauceansine)

[NS] *Delphinium glaucescens*  
 [REF] 212



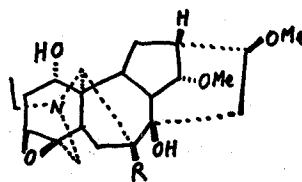
307

[CN] 18-hydroxy-14,10-methylgadesine

[NS] *Consolida orientalis*

[REF] 214

[NC] X-ray analysis



308 R=H (monticamine)

309 R=OH (monticoline)

[NS] *Aconitum monticola*

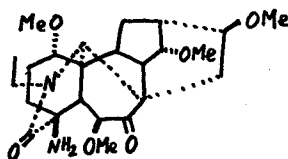
[REF] 215

Additional reference

[REF] 216

[NC] diterpene alkaloids from  
*Delphinium cardiopetalum*

## 2) Synthesis and Reaction



310

[REF] 217

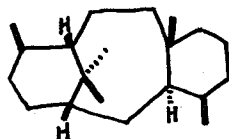
[NC] nitrous acid deamination

## 3) Miscellaneous Section

[REF] 218

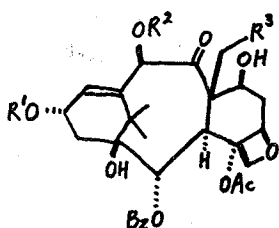
[NC] <sup>13</sup>C NMR spectra

## XIV. TAXANE DERIVATIVES



Taxane

1) Isolation and Structure Determination



311 R<sup>1</sup>=H, R<sup>2</sup>=Ac, R<sup>3</sup>=OH  
(19-hydroxybaccatin IV)

312 R<sup>1</sup>=CO- $\begin{matrix} \text{Ph} \\ | \\ \text{CH} \\ | \\ \text{OH} \end{matrix}$ -CHCHNHCO- $\begin{matrix} \text{Me} & \text{Me} \\ | & | \\ \text{C} & = & \text{CH} \end{matrix}$ ,  
R<sup>2</sup>=R<sup>3</sup>=H  
(10-deacetylcephalomannine)

313 R<sup>1</sup>=COCH $\begin{matrix} \text{Ph} \\ | \\ \text{CH} \\ | \\ \text{OH} \end{matrix}$ NHBz, R<sup>2</sup>=R<sup>3</sup>=H  
(10-deacetyltaxol)

[NS] *Taxus wallichiana*

[REF] 219

[NC] antitumor diterpenoid

314 R<sup>1</sup>=COCH $\begin{matrix} \text{Ph} & \text{Me} & \text{Me} \\ | & | & | \\ \text{CH} & \text{CH} & \text{C} \\ | & & // \\ \text{OH} & & \text{CH} \end{matrix}$ NHCOC=CH,  
R<sup>2</sup>=Ac, R<sup>3</sup>=H

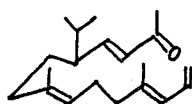
[CN] cephalomannine

[REF] 220

[NC] antileukemic diterpenoid, X-ray analysis

XV. THE OTHERS

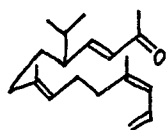
1) Isolation and Structure Determination



315

[REF] 221

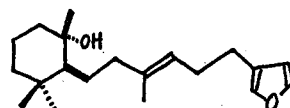
[NC] from cigarette smoke



316

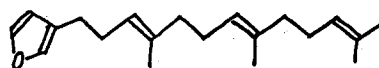
[REF] 221

[NC] from cigarette smoke



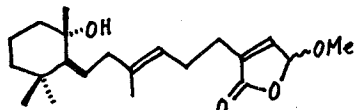
317

[CN] ambliol-A



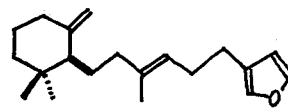
318

[CN] ambliofuran



319

[CN] ambliolide



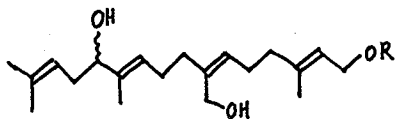
320

[CN] dehydroambliol-A

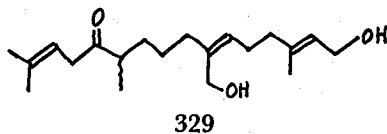
[NS] *Dysidea ambia*

[REF] 45

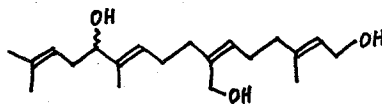
[NC] marine sponge metabolites



321 R=H  
322 R=Ac  
[NC] *Grazielia* species  
[REF] 15

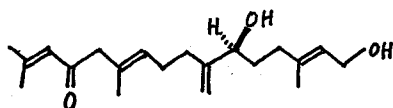


329

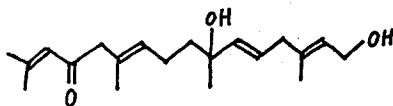


330

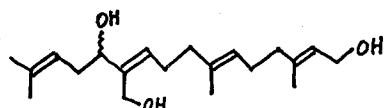
[NS] *Lasiolaena santosii*  
[REF] 20



323

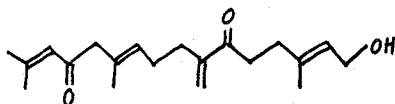


324



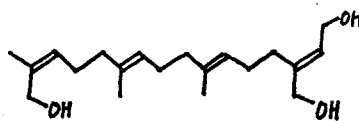
331

[NS] *Zinnia tenuiflora*  
[REF] 225



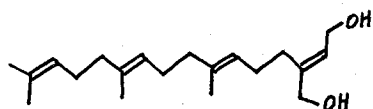
325

[NS] *Cystoseira crinita*  
[REF] 222  
[NC] from brown alga



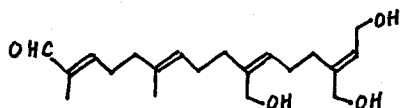
332

[NS] *Bejaranoa semistriata*  
[REF] 226



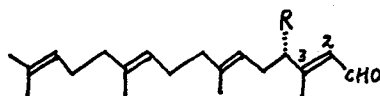
326

[NS] *Kingianthus paradoxus*  
[REF] 223



333

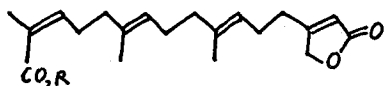
[NS] *Mikania officinalis*



334 R=H

335 R=H (2,3 Z)

336 R=OH



327 R=H

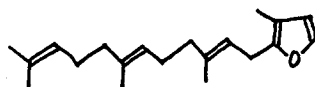
[CN] dimeroperatic acid

328 R=Me

[CN] methyl dimeroperatate

[NS] *Dimerostemma asperatum*

[REF] 224

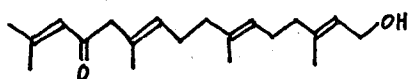


337

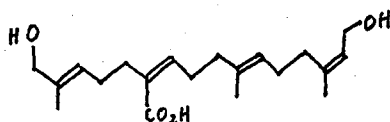
[NS] *Mikania sessilifolia*

[REF] 227

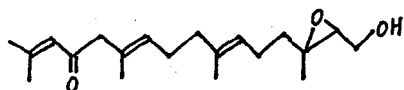




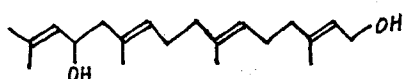
338  
[CN] eleganolone



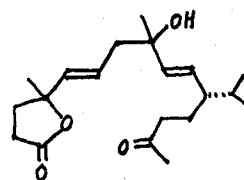
341  
[NS] *Wyethia helenioides*  
[REF] 229



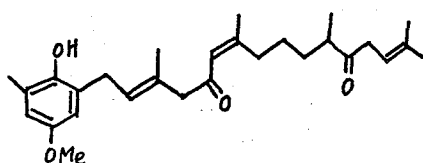
339  
[CN] epoxyeleganolone



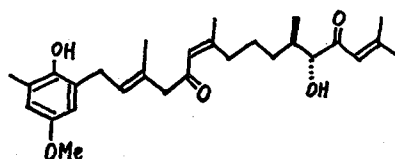
340  
[CN] elegandiol  
[NS] *Cystoseira elegans*  
[REF] 228  
[NC] seasonal variation of  
a cyclic diterpenes



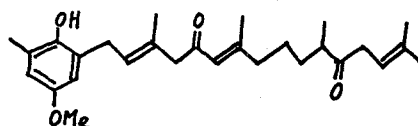
342  
[REF] 230  
[NC] from burley tobacco



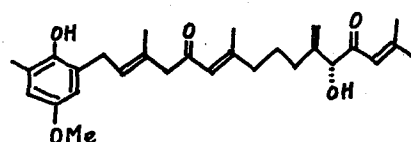
343  
[CN] 5', 12'-dioxohalidrol



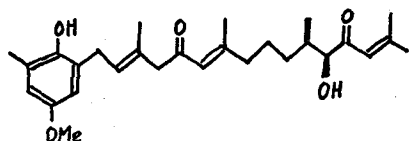
344  
[CN] 12' $\alpha$ -hydroxy-5', 13'-dioxohalidrol



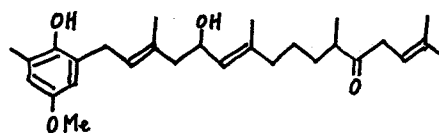
345  
[CN] 5', 12'-dioxoisohalidrol



346  
[CN] 12' $\alpha$ -hydroxy-5', 13'-dioxoisohalidrol

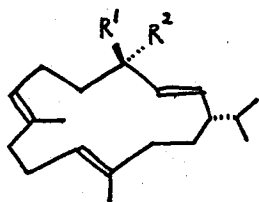


347  
[CN] 12' $\beta$ -hydroxy-5', 13'-  
dioxoisohalidrol

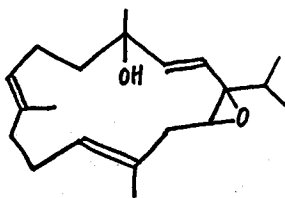


348  
[CN] 5'-hydroxy-12'-oxohalidrol

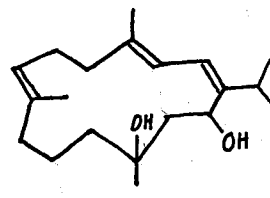
[NS] *Halidrys siliquosa*  
[REF] 231



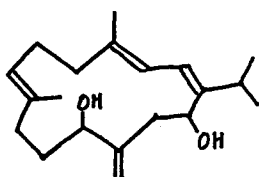
349  $R^1=OH, R^2=Me$   
[CN] thunbegol (isocembrol)  
350  $R^1=Me, R^2=OH$   
[CN] 4-epiisocembrol  
[NS] *Pseudotsuga menziesii*  
[REF] 232



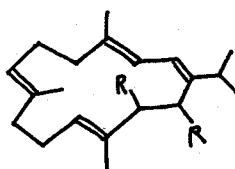
351  
[CN] sarcophytol-C



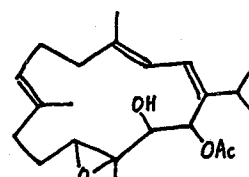
352  
[CN] sarcophytol-D



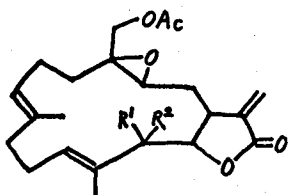
353  
[CN] sarcoahytol-E  
[NS] *Sarcophyton glaucum*  
[REF] 233



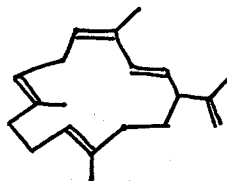
354  $R=H$   
[CN] cembrene-C  
355  $R=OH$   
[CN] sarcophytol-B  
[NS] *Alcyonium flaccidum*  
[REF] 234



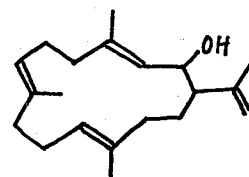
356  
[CN] flaccidoxide  
[NS] *A. flaccidum*  
[REF] 234



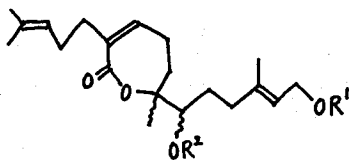
357  $R^1=R^2=H$   
358  $R^1=H, R^2=OH$   
359  $R^1=OH, R^2=H$   
[NS] *Lobophytum crassum*  
[REF] 234



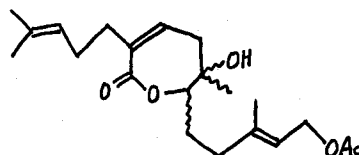
360  
[CN] cembrenene



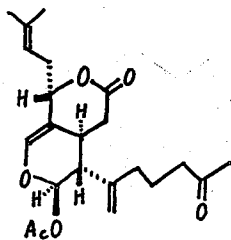
361  
[CN] mayol  
[NS] *Sinuralia mayi*  
[REF] 235



362  $R^1=R^2=H$   
[CN] acanthoaustralide  
363  $R^1=Ac, R^2=H$   
[CN] acanthoaustralide-1-O-acetate  
[NS] *Acanthospermum australe*  
[REF] 236

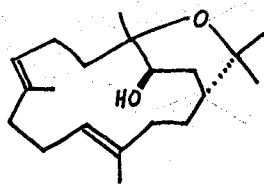


364  
[CN] isoacanthoaustralide-1-O-acetate  
[NS] *A. australe*  
[REF] 236



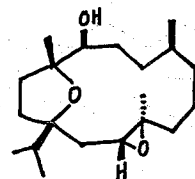
365

[CN] alcyonolide  
[NS] *Akyonimu* species  
[REF] 237



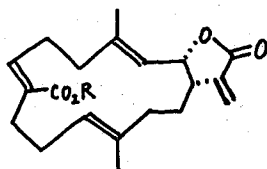
366

[CN] decaryiol  
[NS] *Sarcophyton decaryi*  
[REF] 238



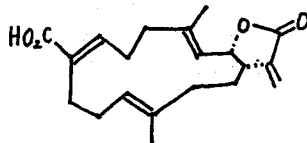
367

[CN] incensole oxide  
[REF] 239  
[NC] X-ray analysis



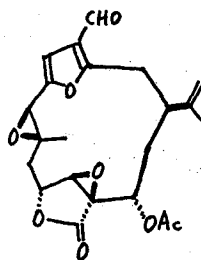
368

[CN] lobohedleolide  
[NS] *Lobophytum hedleyi*  
[REF] 240  
[NC] X-ray analysis;  
growth inhibition  
of the Hella cells



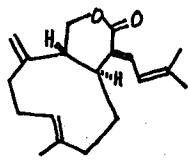
369

[CN] (7Z)-lobohedleolide  
[NS] *L. hedleyi*  
[REF] 240



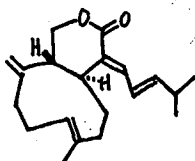
370

[CN] lophotoxin  
[NS] *Lophogorgia alba*,  
*L. cuspidata*, *L. rigida*,  
*L. chilensis*  
[REF] 241  
[NC] new neuromuscular  
toxin



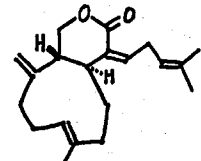
371

[CN] coraxeniolide-A



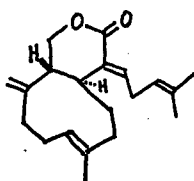
372

[CN] coraxeniolide-B



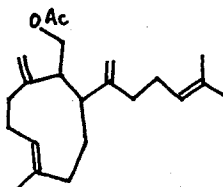
373

[CN] coraxeniolide-C



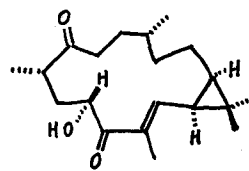
374

[CN] coraxeniolide-C'



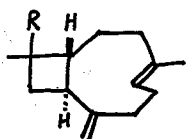
375

[CN] corabohcin  
[NS] *Corallium* species  
[REF] 242



376

[CN] crotonitenone  
[NS] *Croton nitens*  
[REF] 243

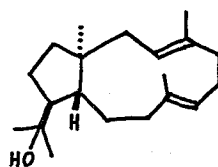


377 R = CH<sub>2</sub>CH<sub>2</sub>CH—CMe<sub>2</sub>

378 R = CH<sub>2</sub>CH<sub>2</sub>CH—CMe<sub>2</sub>  
                                   OH Cl

[NS] *Nephtea chabrolii*

[REF] 244

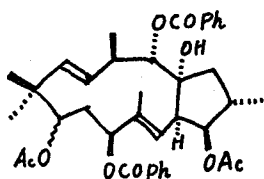


379

[CN] 18-hydroxy-3,7-dolablladiene

[NS] *Dictyota dichotoma*

[REF] 245

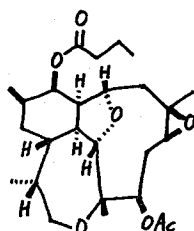


380

[NC] euphornin

[NS] *Euphorbia maddenii*

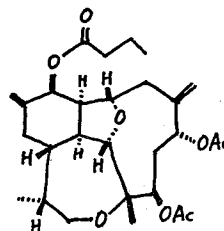
[REF] 246



381

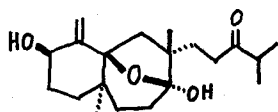
[NS] *Briareum asbestinum*

[REF] 247



382

[NC] from toxic extracts

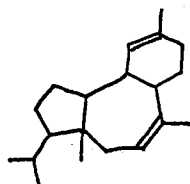


383

[CN] linearol

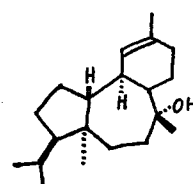
[NS] *Dictyota linearis*

[REF] 248



384

[CN] sphaerodiene



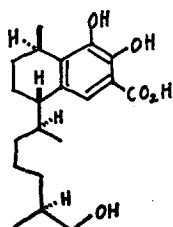
385

[CN] presphaerol

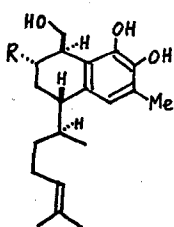
[NS] *Sphaerococcus coronopifolius*

[REF] 249

[NC] structure 385  
 (reassigned by  
 X-ray analysis)



386



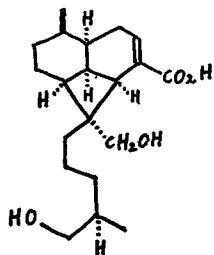
387 R = H

388 R = OH

[NS] *Eremophila* species

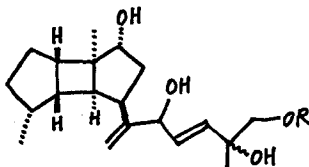
[REF] 250, 251

[NC] X-ray analysis



389

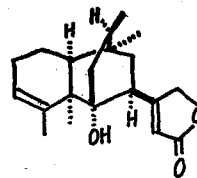
[NS] *Eremophila decipiens*  
[REF] 252  
[NC] chemical degradation



390 R=H, 391 R=Ac

[NS] *Stoechospermum marginatum*

[REF] 253



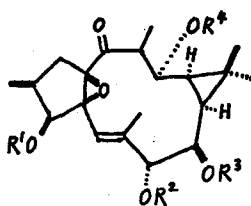
392

[CN] tricyclosolidagolactone

[NS] *Solidago altissima*

[REF] 254

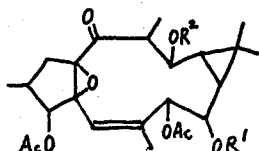
[NC] X-ray analysis



	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>
393	Ac	Ac	benzoate	Ac
394	Ac	Ac	angelate	Ac
395	Ac	Ac	tigliate	Ac
396	H	Ac	benzoate	Ac
397	H	Ac	angelate	Ac
398	H	H	tigliate	Ac
399	H	H	H	Ac
400	H	H	H	H
401	Ac	Ac	Ac	Ac

[NS] *Euphorbia kamerunica*

[REF] 255

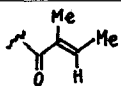


R<sup>1</sup>

R<sup>2</sup>

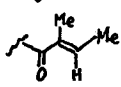
402

Ac



403

H



404

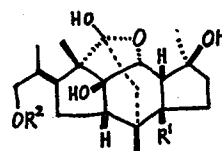
Ac

Ac

[NS] *Euphorbia kamerunica*

[REF] 256

[NC] from the cytotoxic fraction



405 R<sup>1</sup>=R<sup>2</sup>=H

[CN] cinncassiol D<sub>1</sub>

406 R<sup>1</sup>=H, R<sup>2</sup>=β-D-glc·pyr-

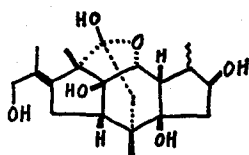
[CN] cinncassiol D<sub>1</sub> glucoside

407 R<sup>1</sup>=OH, R<sup>2</sup>=H

[CN] cinncassiol D<sub>2</sub>

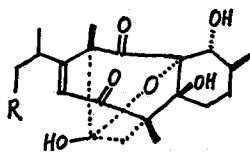
408 R<sup>1</sup>=OH, R<sup>2</sup>=β-D-glc·pyr-

[CN] cinncassiol D<sub>2</sub> glucoside



409

[CN] cinnassiol D<sub>3</sub>  
 [NS] *Cinnamomi cortex*  
 [REF] 257  
 [NC] from the fraction exhibiting anti-complement activity; X-ray analyses

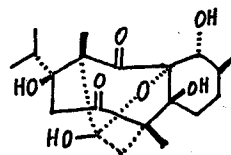


410 R = O-β-D-glc·pyr

[CN] cinnassiol C<sub>1</sub>  
 glucoside

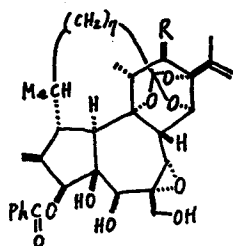
411 R = H

[CN] cinnassiol C<sub>2</sub>



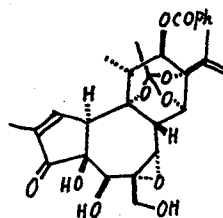
412

[CN] cinnassiol C<sub>3</sub>  
 [NS] *Cinnamomi cortex*  
 [REF] 258  
 [NC] from the fraction exhibiting anticomplement activity



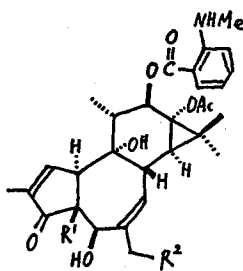
413 R = OAc  
 [CN] linifolin A

414 R = H  
 [CN] linifolin B  
 [NS] *Pimelea linifolia*  
 [REF] 259  
 [NC] fish toxin



415

[CN] genkwadaphnin  
 [NS] *Daphne genkwa*  
 [REF] 260  
 [NC] new antileukemic principle



416 R<sup>1</sup> = H, R<sup>2</sup> = OH

[CN] sapintoxin B

417 R<sup>1</sup> = R<sup>2</sup> = H

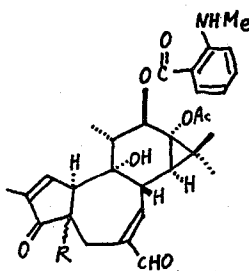
[CN] sapintoxin C

418 R<sup>1</sup> = R<sup>2</sup> = OH

[CN] sapintoxin D

[NS] *Sapium indicum*

[REF] 261, 262



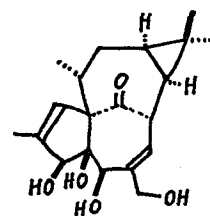
419 R = β-H

420 R = α-H

[NS] *Sapium indicum*

[REF] 263

[NC] first natural tiglianes to exhibit a C-20 aldehyde

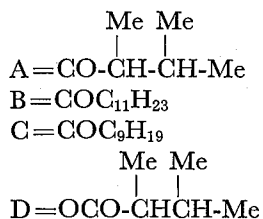
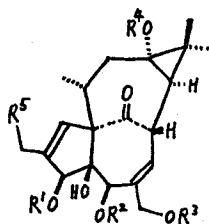


421

[CN] ingenol

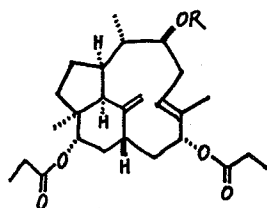
[NS] *Euphorbia ingens*

[REF] 264

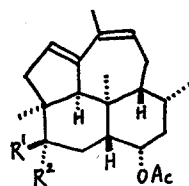


- 422 R<sup>2</sup>=R<sup>3</sup>=R<sup>5</sup>=H, R<sup>1</sup>=A, R<sup>4</sup>=B  
423 R<sup>1</sup>=R<sup>3</sup>=R<sup>5</sup>=H, R<sup>2</sup>=A, R<sup>4</sup>=B  
424 R<sup>1</sup>=R<sup>2</sup>=R<sup>5</sup>=H, R<sup>3</sup>=A, R<sup>4</sup>=B  
425 R<sup>1</sup>=A, R<sup>2</sup>=R<sup>3</sup>=R<sup>5</sup>=H, R<sup>4</sup>=C  
426 R<sup>1</sup>=R<sup>4</sup>=A, R<sup>2</sup>=R<sup>3</sup>=H, R<sup>5</sup>=D  
427 R<sup>1</sup>=R<sup>3</sup>=H, R<sup>2</sup>=R<sup>4</sup>=A, R<sup>5</sup>=D  
428 R<sup>1</sup>=R<sup>2</sup>=H, R<sup>3</sup>=R<sup>4</sup>=A, R<sup>5</sup>=D

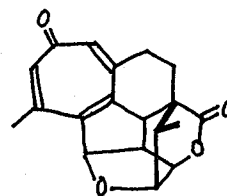
[NS] *Euphorbia cyparissias*  
[REF] 265  
[NC] highly irritant activity



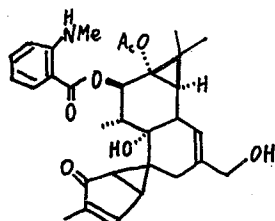
- 429 R=COEt  
430 R=COMe  
[NS] *Nasutitermes* species  
[REF] 266  
[NC] termite soldier  
defense secretion



- 431 R<sup>1</sup>=R<sup>2</sup>=O  
432 R<sup>1</sup>=OAc, R<sup>2</sup>=H  
433 R<sup>1</sup>=H, R<sup>2</sup>=OAc  
[NS] *Bulbitermes singaporensis*  
[REF] 267  
[NC] defense secretion of the  
nasute termite



- 434  
[CN] harringtonolide  
[NS] *Cephalotaxus harringtonia*  
[REF] 84  
[NC] podolactone-type  
plant growth  
inhibition

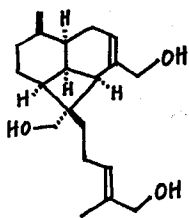


- 435  
[CN] sapintoxin A  
[NS] *Sapium indicum*  
[REF] 268  
[NC] irritant compound

Additional references

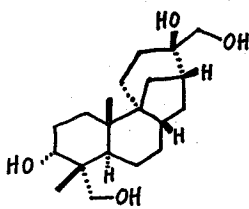
- [REF] 12  
[NC] cembrane diterpenoids from oleoresin of  
*Picea-ajanensis*  
[REF] 269, 270  
[NC] phytochemical cultivation of Brazilian  
velloziaceae  
[REF] 271  
[NC] diterpenoids of oleoresin of far east *Abies*  
species  
[REF] 272  
[NC] new diterpenic acetate from a *Sideritis pusilla*

2) Synthesis and Reaction



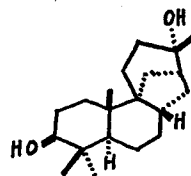
436

[CN] (+)trihydroxydecipadiene  
[REF] 273  
[NC] total synthesis



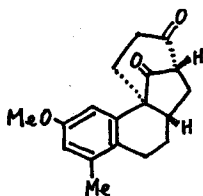
437

[CN] (+)aphidicolin  
[REF] 274, 275  
[NC] total synthesis



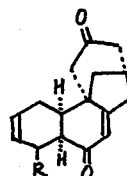
438

[CN] (+)maritimol  
[REF] 276  
[NC] total synthesis



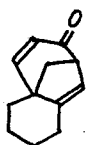
439

[REF] 277  
[NC] synthesis of the basic skeleton of aphidicolan-type diterpenes

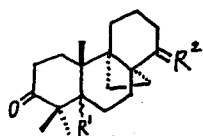


440 R=H, 441 R=OAc

[REF] 278  
[NC] synthesis of the ring skeleton of aphidicolin and the related natural products

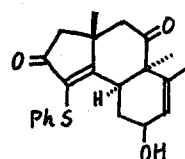


442



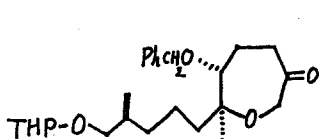
443 R<sup>1</sup>= $\alpha$ -H, R<sup>2</sup>=O  
444 R<sup>1</sup>= $\beta$ -H, R<sup>2</sup>= $\alpha$ -OH,  $\beta$ -H

[REF] 279  
[NC] preliminary studies on the synthesis of aphidicolin



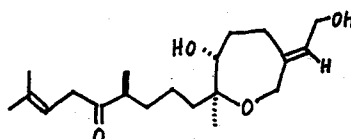
445

[REF] 280  
[NC] synthesis of a potential intermediate directed towards cyathins



446

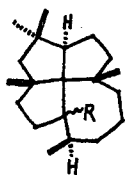
[REF] 281  
[NC] stereoselective synthesis of a key intermediate for the total synthesis of (+)zoapatanol



447

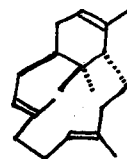
[CN] (+)zoapatanol  
[REF] 282  
[NC] total synthesis



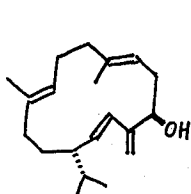


448 R= $\alpha$ -H  
[CN] laurenane

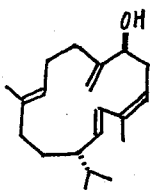
449 R= $\beta$ -H  
[CN] 1 $\beta$ H-laurenane  
[REF] 283



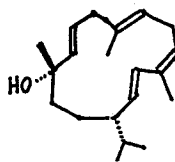
450  
[REF] 284  
[NC] synthesis of a geometrical isomer of anhydroverticillol



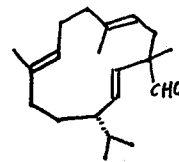
451



452

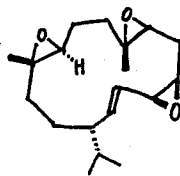


453



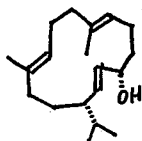
454

[REF] 285  
[NC] photooxidation products of cembrene



455

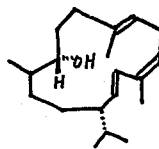
[REF] 286  
[NC] complete epoxidation product of cembrene; X-ray analysis



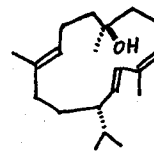
456

[CN] cembranol

453

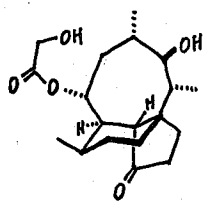


457



458

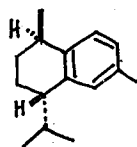
[REF] 287  
[NC] reduction products of 11S,12S-epoxycembrene or 7S,8S-epoxycembrene by LiAlH<sub>4</sub>; growth inhibition of red wheat



459

[REF] 288

[NC] chemical conversion from a pleuromutilin derivative

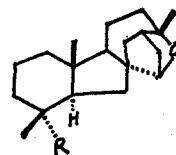


460

[CN] (1*R*,4*S*)-(-)-calamenene

[REF] 289

[NC] synthesis from dihydroxyserrulatic acid



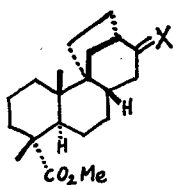
461 R=Me

462 R=CH<sub>2</sub>OCOPh

463 R=CO<sub>2</sub>Me

[REF] 290

[NC] chemical conversion (*in vitro* biosynthetic studies); X-ray analysis

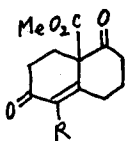


464 X=α-Me, β-OMe

465 X=α-OMe, β-Me

[REF] 291

[NC] cyclization products of methyl isopimarate and pimarate



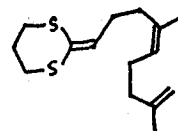
466 R=H

467 R=Me

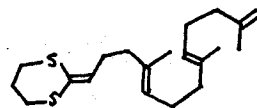
[CN] Wieland-Mischer ketone analogues

[REF] 292

[NC] intermediates in the synthesis of bruceantin



468



469

[REF] 293

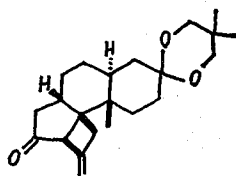
[NC] synthesis of *Z*- and *E*-polyunsaturated isoprenoids

Additional reference

[REF] 294

[NC] regiospecific and stereoselective 3α-hydroxylation of fusicoccin derivatives

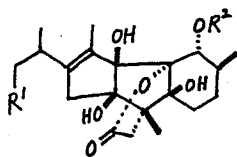
3) Miscellaneous Section



470

[REF] 295

[NC] X-ray analysis of a stemodin intermediate 470



471 R<sup>1</sup>=H, R<sup>2</sup>=Ac

472 R<sup>1</sup>=R<sup>2</sup>=H

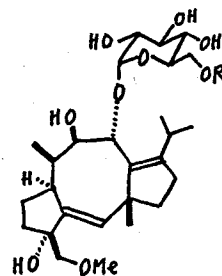
473 R<sup>1</sup>=OH, R<sup>2</sup>=H

474 R<sup>1</sup>=OAc, R<sup>2</sup>=H

475 R<sup>1</sup>=O-glc(Ac), R<sup>2</sup>=H

[REF] 296

[NC] EI mass spectra of *Cassia* diterpenes



476 R=Me

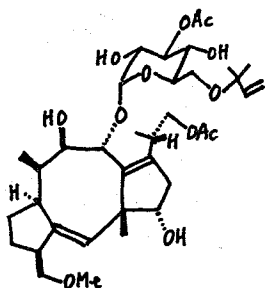
[CN] cotylenin-E

477 R=H

[CN] cotylenin-J

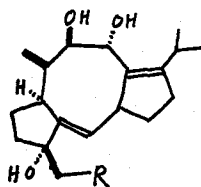
[REF] 297

[NC] germination-stimulating activity



478

[CN] fusicoccin-A  
[REF] 297  
[NC] germination-stimulating activity



479 R=OMe

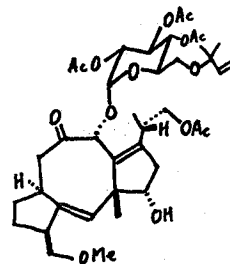
[CN] cotylenol

480 R=OH

[CN] 16-O-demethylcotylenol

[REF] 297

[NC] germination-stimulating activity

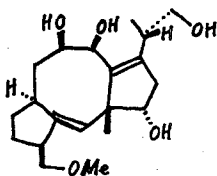


481

[CN] fusicoccin triacetate

[REF] 298

[NC] structure revision (481); preparation of 482

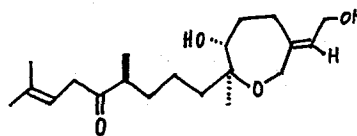


482

[CN] 9-epifusicoccin aglycone

[REF] 298

[NC] structure revision (481); preparation of 482

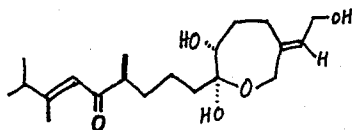


446

[CN] zoapatanol

[REF] 299

[NC] <sup>13</sup>C NMR studies

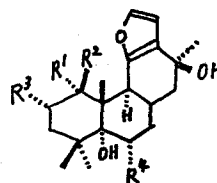


483

[CN] montanol

[REF] 299

[NC] <sup>13</sup>C NMR studies



484 R<sup>1</sup>=R<sup>3</sup>=OAc, R<sup>2</sup>=R<sup>4</sup>=H

485 R<sup>1</sup>, R<sup>2</sup>=O, R<sup>3</sup>=H, R<sup>4</sup>=OAc

486 R<sup>1</sup>, R<sup>2</sup>=O, R<sup>3</sup>=H, R<sup>4</sup>=OH

487 R<sup>1</sup>, R<sup>4</sup>=OH, R<sup>2</sup>=R<sup>3</sup>=H

[RER] 300

[NC] <sup>13</sup>C NMR studies of cassane diterpenoids; stereochemistry of the caesalpins

Additional reference

[REF] 301

[NC] computer-assisted structural interpretation of <sup>13</sup>C NMR spectral data

XVI. REVIEW ARTICLES

[REF] 302

[NC] irritant and defense substances

[REF] 303

[NC] terpenoid metabolites of mushrooms

[REF] 304

[NC] naturally occurring gibberellins (in Japanese)

[REF] 305

[NC] biologically active glucosides produced by fungi (in Japanese)

[REF] 306

[NC] fungal elicitors of the phytoalexin response

[REF] 307

[NC] chemistry of new natural sweet-principles (in Japanese)

[REF] 308

[NC] cocarcinogenesis and tumor promoters

[REF] 309

[NC] Chinese medicinal plants (in Japanese)

[REF] 310

[NC] decomposition of  $\alpha$ -diazo ketones

[REF] 311

[NC] singlet oxygen in organic synthesis

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