Review

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Chemical and pharmacological research on the plants from genus *Ajuga*

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Abstract: The genus *Ajuga*, a member of the Lamiaceae family, is comprised of more than 300 species of annual and perennial herbaceous flowering plants mainly distributed throughout the temperate regions of Asia, Europe, Australia, North America and Africa. These plants are used as folk medicines effective for rheumatic fevers, dysentery, malaria, hypertension, diabetes and gastrointestinal disorders, as well as anthelmintic, astringent, febrifuge diuretic, antifungal and anti-inflammatory agents. A variety of constituents has been isolated from these plants. This review summarizes the phytochemical progress of the genus *Ajuga* and lists the compounds isolated up to 2014.

Keywords: *Ajuga*; biological activity; chemical constituents; *Lamiaceae*; phytoecdysteroids; terpenoi.

Introduction

The genus *Ajuga*, a member of the Lamiaceae family, is comprised of more than 300 species of annual and perennial herbaceous flowering plants mainly distributed throughout the temperate regions of Asia, Europe, Australia, North America and Africa. These species have been used as common house plants and are called bugle or bugleweed. They are mainly characterized by the color and shape of the flower. For example, the flower of *Ajuga reptans* is somewhat tall and blue, while that of *Ajuga decumbens* is short and purple. Many of these plants are of medicinal importance and are traditionally used as remedies for rheumatic fevers, dysentery, malaria, hypertension, diabetes and gastrointestinal disorders, as well as anthelmintic, astringent, febrifuge diuretic, antifungal and anti-inflammatory agents [1]. The genus *Ajuga* has attracted attention since the report in 1976 that *Ajuga remota* grown in Kenya is not attacked by African armyworms and contains three moderately strong antifeedants [2]. Since then, reports of the isolation of neoclerodanes and phytoecdysteroids, as the insect allelochemicals responsible for antifeedant activity from this genus, have appeared [3]. Several species of this genus have been chemically studied and a series of bioactive metabolites, including phytoecdysteroids, diterpenoids and iridoids have been isolated and characterized. Biological investigations demonstrate that some of these compounds display antibacterial [4], antifungal [5], antiplasmodial [6], cytotoxic, antitumor promoting [7], vasoconstricting [8], insect molting inhibitory, insect antifeeding [9] and enzyme-inhibitory [10] activities. This review summarizes phytochemical progress of the genus *Ajuga* covering the literature up to 2014. In addition, some biological activities of compounds obtained from this genus are also listed.

Chemical constituents

There have been many phytochemical investigations on the isolation of constituents from the *Ajuga* genus. This has resulted in the isolation and characterization of a series of secondary metabolites, including phytoecdysteroids, sesquiterpenoids, diterpenoids, triterpenoids,

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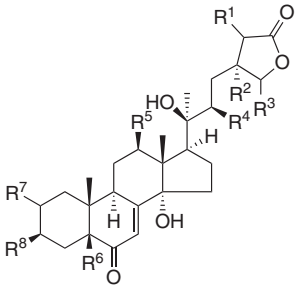
Table 1 Steroids 1: Phytoecdysteroids.

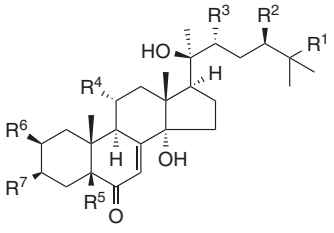
No.	Name	Source	Part	Ref.
1	Cyasterone	*A. decumbens*	whole plant	[11]
		A. turkestanica	leaf	[12]
		A. iva	aerial part	[13–15]
		A. chia	leaf, stem	[16]
		A. chamaepitys	whole plant	[17]
		A. multiflora	aerial part	[18]
		A. taiwanensis	whole plant	[19]
		A. nipponensis	aerial part	[20]
		A. macrosperma var. *breviflora*	root	[21]
2	Ecdysterone	*A. decumbens*	whole plant	[11]
		A. nipponensis	whole plant	[11, 22]
		A. turkestanica	leaf	[12]
		A. iva	aerial part	[13–15]
		A. chamaepitys	whole plant	[17]
		A. multiflora	aerial part	[18]
		A. macrosperma var. *breviflora*	root	[21]
		A. reptans	whole plant	[23–25]
		A. remota	leaf, root	[26]
3	Ajugalactone	*A. reptans*	whole plant	[25]
		A. turkestanica	root	[27]
4	22-Acetylcyasterone	*A. turkestanica*	root	[28]
5	Turkesterone	*A. turkestanica*	root	[29]
6	Ajugasterone A	*A. nipponensis*	whole plant	[22]
		A. reptans	whole plant	[23, 25]
7	Ajugasterone B	*A. reptans*	whole plant	[25]
		A. turkestanica	root	[29]
		A. iva	whole plant	[30]
8	Ajugasterone C	*A. nipponensis*	aerial part	[20]
		A. japonica	leaf	[31]
9	Ajugasterone D	*A. nipponensis*	whole plant	[22]
10	Stachysterone D	*A. nipponensis*	whole plant	[22]
11	Makisterone A	*A. iva*	whole plant	[14, 32]
		A. macrosperma var. *breviflora*	root	[21]
12	29-Norcyasterone	*A. reptans*	whole plant	[23–25]
13	29-Norsengosterone	*A. reptans*	whole plant	[23–25]
14	2-Acetyl-29-norcyasterone	*A. reptans*	whole plant	[33]
15	3-Acetyl-29-norcyasterone	*A. reptans*	whole plant	[33]
16	Sengosterone	*A. reptans*	whole plant	[25]
17	22-Dehydro-12-hydroxycyasterone	*A. reptans* var. *atropurperea*	aerial part	[34]
18	22-Dehydro-12-hydroxysengosterone	*A. reptans* var. *atropurperea*	aerial part	[34]
19	22-Dehydro-12-hydroxy-29-nor-cyasterone	*A. reptans* var. *atropurperea*	aerial part	[34]
20	22-Dehydro-12-hydroxy-29-nor-sengosterone	*A. reptans* var. *atropurperea*	aerial part	[34]
21	Reptansterone	*A. reptans* var. *atropurperea*	root	[35]
22	28-*epi*-Sengosterone	*A. reptans* var. *atropurperea*	root	[35]
23	5,29-Dihydroxycapitasterone	*A. reptans* var. *atropurperea*	root	[35]
24	2-Dehydroajugalactone	*A. reptans* var. *atropurperea*	root	[35]
25	3-Dehydroajugalactone	*A. reptans* var. *atropurperea*	root	[35]
26	Ponasterone A	*A. remota*	leaf, root	[26]
27	24,28-Dehydromakisterone A	*A. iva*	whole plant	[14]
28	22-Oxocyasterone	*A. iva*	whole plant aerial	[14]
	22-Dehydrocyasterone	*A. nipponensis*	part	[20]
29	24,25-Dehydroprecyasterone	*A. iva*	whole plant	[14]
		A. reptans var. *reptans*	whole plant	[36]
30	20-Hydroxyecdysone 22-acetate	*A. nipponensis*	aerial part	[20]
		A. reptans	whole plant	[37]
31	20-Hydroxyecdysone 25-acetate (Viticosterone E)	*A. reptans*	whole plant	[37]

Table 1 (continued)

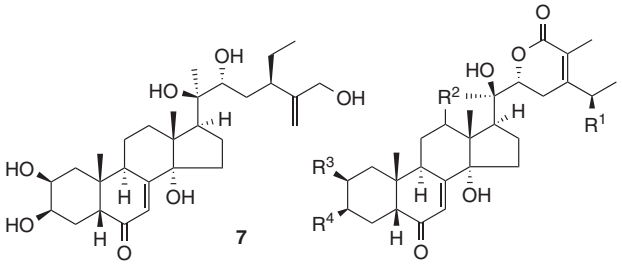
No.	Name	Source	Part	Ref.
32	Ajusterone	*A. pseudoiva*	leaf	[38]
33	Ajugalide-E	*A. taiwanensis*	whole plant	[19]
34	Isocyasterone	*A. taiwanensis*	whole plant	[19]
35	24-Hydroxycyasterone	*A. iva*	whole plant	[30]
36	22-Dehydrocyasterone 2-glucopyranoside	*A. nipponensis*	aerial part	[20]
37	Ajugacetalsterone A	*A. nipponensis*	aerial part	[20]
38	Ajugacetalsterone B	*A. nipponensis*	aerial part	[20]
39	Ajugacetalsterone C	*A. macrosperma* var. *breviflora*	root	[21]
40	Ajugacetalsterone D	*A. macrosperma* var. *breviflora*	root	[21]
41	Breviflorasterone	*A. macrosperma* var. *breviflora*	root	[21]
		A. reptans var. *reptans*	whole plant	[36]
42	20-Hydroxyecdysone 2-acetate	*A. macrosperma* var. *breviflora*	root	[21]
43	20-Hydroxyecdysone 3-acetate	*A. macrosperma* var. *breviflora*	root	[21]
44	Reptanslactone A	*A. reptans* var. *reptans*	whole plant	[36]
45	Reptanslactone B	*A. reptans* var. *reptans*	whole plant	[36]
46	Sendreisterone	*A. reptans* var. *reptans*	whole plant	[36]



	R^1	R^2	R^3	R^4	R^5	R^6	R^7	R^8
1	α-Me	H	α-Me	OH	H	H	β-OH	OH
4	α-Me	H	α-Me	OAc	H	H	β-OH	OH
12	α-Me	H	H	OH	H	H	β-OH	OH
13	α-Me	H	H	OH	H	OH	β-OH	OH
14	α-Me	H	H	OH	H	H	β-OAc	OH
15	α-Me	H	H	OH	H	H	β-OH	OAc
16	α-Me	H	α-Me	OH	H	OH	β-OH	OH
17	α-Me	H	α-Me	=O	OH	H	β-OH	OH
18	α-Me	H	α-Me	=O	OH	OH	β-OH	OH
19	α-Me	H	H	=O	OH	H	β-OH	OH
20	α-Me	H	H	=O	OH	OH	β-OH	OH
22	α-Me	H	β-Me	OH	H	OH	β-OH	OH
28	α-Me	H	α-Me	=O	H	H	β-OH	OH
33	α-Me	H	α-Me	OAc	H	H	α-OH	OH
34	β-Me	H	α-Me	OH	H	H	β-OH	OH
35	α-Me	OH	α-Me	OH	H	H	β-OH	OH
36	α-Me	H	α-Me	=O	H	H	β-O-Glc	OH



	R^1	R^2	R^3	R^4	R^5	R^6	R^7
2	OH	H	OH	H	H	OH	OH
5	OH	H	OH	OH	H	OH	OH
6	OH	H	OH	H	OH	OH	OH
8	H	H	OH	OH	H	OH	OH
11	OH	Me	OH	H	H	OH	OH
26	H	H	OH	H	H	OH	OH
27	OH	$=CH_2$	OH	H	H	OH	OH
30	OH	H	OAc	H	H	OH	OH
31	OAc	H	OH	H	H	OH	OH
32	OH	H	OH	H	H	OH	OH
42	OH	H	OH	H	H	OAc	OH
43	OH	H	OH	H	H	OH	OAc



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	R^1	R^2	R^3	R^4
3	H	=O	OH	OH
24	H	=O	=O	OH
25	H	=O	OH	=O
29	OH	H	OH	OH

Figure 1 Steroids 1: Phytoecdysteroids.

Table 2 Steroids 2: Withanolides.

No.	Name	Source	Part	Ref.
47	Ajugin	*A. parviflora*	whole plant	[40]
48	Ajugin A	*A. parviflora*	whole plant	[41]
49	Ajugin B	*A. parviflora*	whole plant	[41]
50	Ajugin C	*A. parviflora*	whole plant	[42]
51	Ajugin D	*A. parviflora*	whole plant	[42]
52	Ajugin E	*A. parviflora*	whole plant	[43]
53	Ajugin F	*A. parviflora*	whole plant	[43]
54	3,14,17,20,28-Pentahydroxy-1-oxo-(20*R*,22*R*)-witha-5,24-dienolide	*A. parviflora*	whole plant	[44]
55	3,17,20-Trihydroxy-1-oxo-(20*S*,22*R*)-witha-5,14,24-trienolide	*A. parviflora*	whole plant	[45]
56	28-Hydroxy-14,20-epoxy-1-oxo-(22*R*)-witha-2,5,24-trienolide	*A. parviflora*	whole plant	[45]
57	Coagulin-J	*A. parviflora*	whole plant	[44]
58	Bracteosin A	*A. bracteosa*	whole plant	[46]
59	Bracteosin B	*A. bracteosa*	whole plant	[46]
60	Bracteosin C	*A. bracteosa*	whole plant	[46]

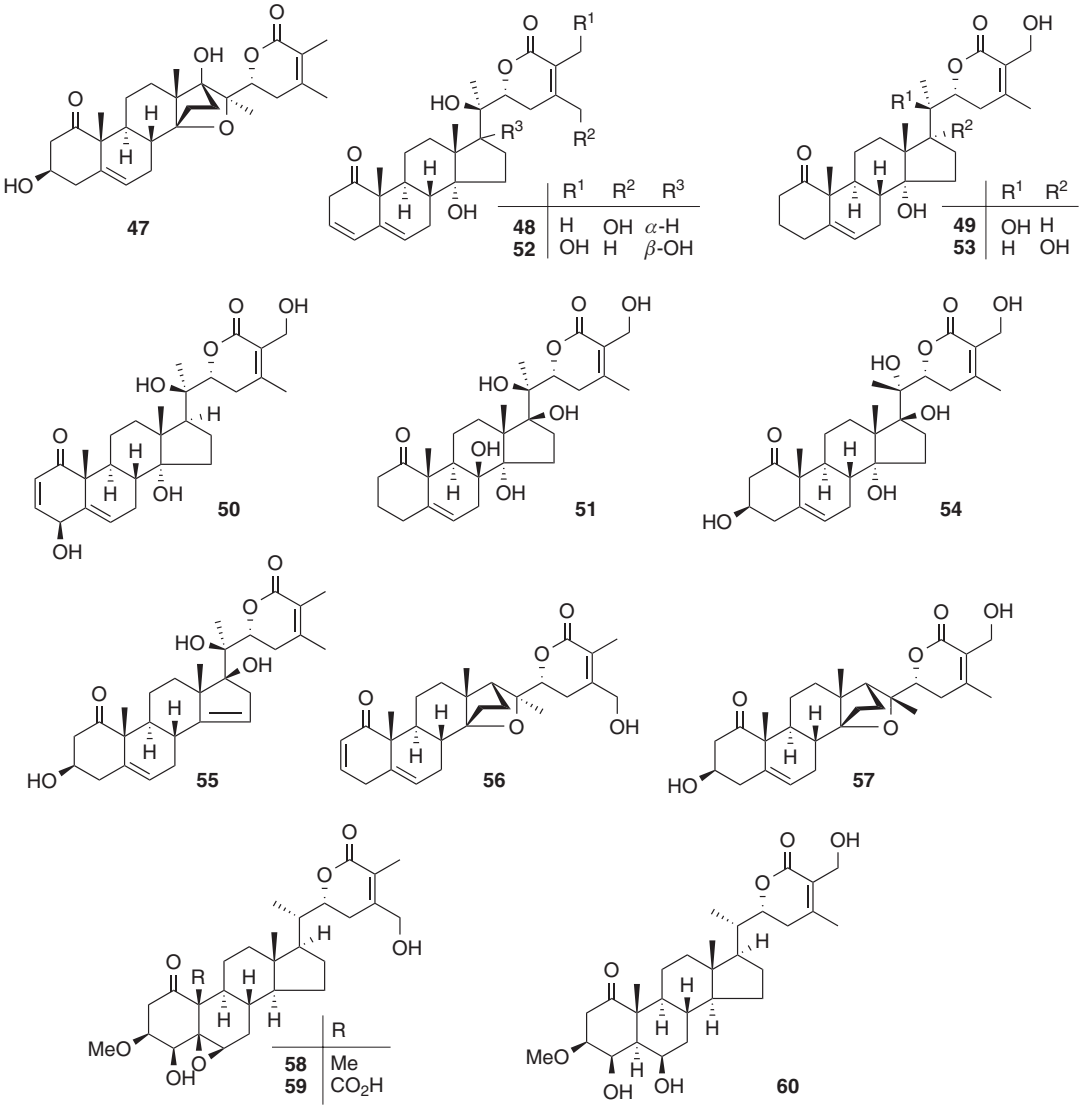


Figure 2 Steroids 2: Withanolides.

Table 3 Steroids 3: Other steroids.

No.	Name	Source	Part	Ref.
61	Clerosterol	*A. reptans*	whole plant	[47]
		A. pseudoiva	leaf	[48]
		A. relicta	whole plant	[49]
62	Clerosterol 3β-*O*-(β-D-glucopyranoside)	*A. pseudoiva*	leaf	[50]
63	Mighavide (3-*O*-Butanoylclerosterol)	*A. pseudoiva*	leaf	[48, 50]
64	3-*O*-β-D-Glucopyranosyl-stigmasta-5,25-diene	*A. chamaepitys* ssp. *laevigata*	whole plant	[51]
65	Stigmasterol	*A. taiwanensis*	whole plant	[19]
66	Stigmasterol 3-*O*-β-D-glucopyranoside	*A. taiwanensis*	whole plant	[19]
67	22,23-Didehydroclerosterol	*A. reptans*	whole plant	[47]
68	β-sitosterol	*A. relicta*	whole plant	[49]
		A. taiwanensis	whole plant	[19]
69	β-sitosterol 3-*O*-β-D-glucopyranoside	*A. decumbens*	whole plant	[52]
70	Ajugasalicioside A	*A. salicifolia*	aerial part	[53]
71	Ajugasalicioside B	*A. salicifolia*	aerial part	[53]
72	Ajugasalicigenin	*A. salicifolia*	aerial part	[54]
73	Ajugasalicioside C	*A. salicifolia*	aerial part	[53]
74	Ajugasalicioside D	*A. salicifolia*	aerial part	[53]
75	Ajugasalicioside E	*A. salicifolia*	aerial part	[53]
76	Ajugasalicioside F	*A. salicifolia*	aerial part	[54]
77	Ajugasalicioside G	*A. salicifolia*	aerial part	[54]
78	Ajugasalicioside H	*A. salicifolia*	aerial part	[54]
79	(24*S*)-24-Ethyl-11α-hydroxycholesta-5,25-dien-1-one	*A. relicta*	whole plant	[49]
80	(24*S*)-24-Ethyl-7α-hydroxycholesta-5,25-dien-3-one	*A. relicta*	whole plant	[49]
81	Ergosterol 5,8-endoperoxide	*A. remota*	aerial part	[55]

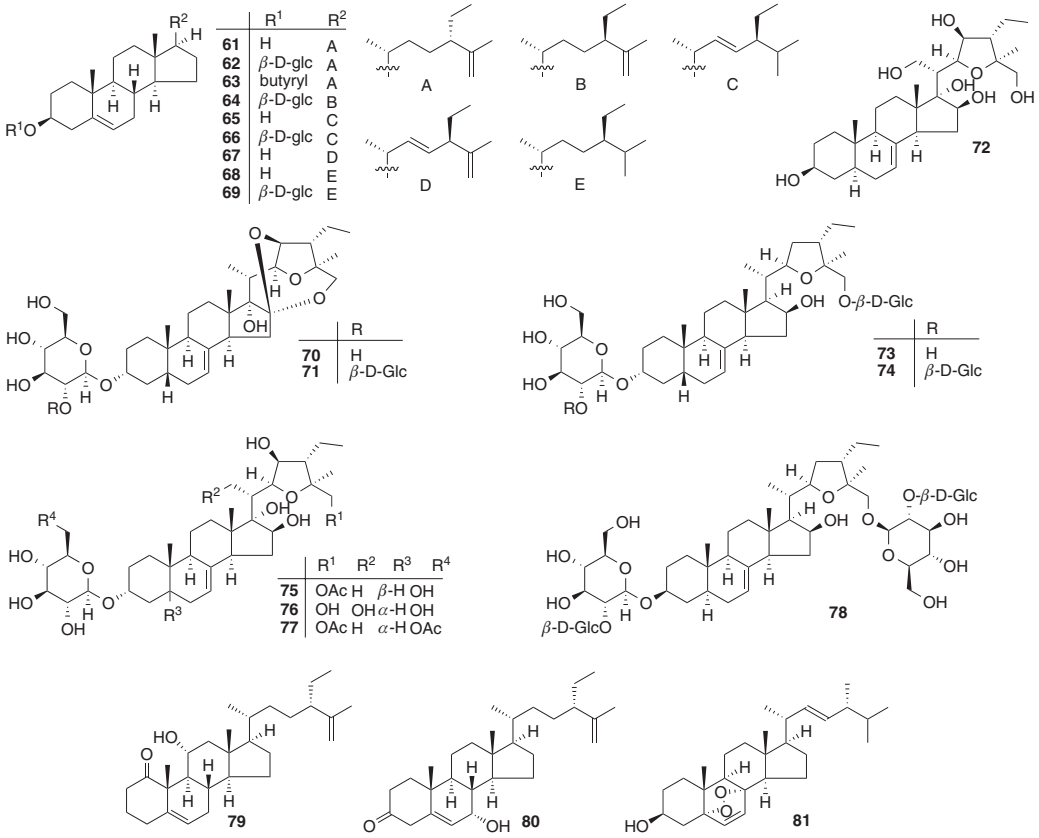


Figure 3 Steroids 3: other steroids.

Table 4 Triterpenoids.

No.	Name	Source	Part	Ref.
82	Betulinic acid	*A. macrosperma*	aerial part	[56]
83	3-*epi*-Betulinic acid	*A. macrosperma*	aerial part	[56]
84	Oleanolic acid	*A. relicta*	whole plant	[49]
85	3-*O*-Acetyloleanolic acid	*A. relicta*	whole plant	[49]
86	α-Amyrin	*A. chamaepitys* ssp. *laevigata*	whole plant	[51]
87	β-Amyrin	*A. chamaepitys* ssp. *laevigata*	whole plant	[51]
88	Ursolic acid	*A. chamaepitys* ssp. *laevigata*	whole plant	[51]

iridoids, withanolides and some other compounds. Phytoecdysteroids, the characteristic components of *Ajuga* plants, are discussed first.

Steroids

Phytoecdysteroids (Table 1, Figure 1)

Phytoecdysteroids are widespread in the genus *Ajuga*. These compounds display interesting physiological activities such as insect molting activity and other hormonal functions involving regeneration, metamorphosis, reproduction and differentiation in all arthropods. These compounds play important roles for defense against phytophagous insects. They also display antiulcer, antirheumatic, insulin regulation and diuretic or tonic activities in mammals [34]. Some of the successful applications of plants in folk medicine can be explained by the occurrence of phytoecdysteroids.

Compounds **1** and **2** are usually the most abundant phytoecdysones in the genus *Ajuga*, and they were reported in *A. decumbens, Ajuga incisa, Ajuga turkestanica, Ajuga iva, Ajuga nipponensis, Ajuga chia*, *Ajuga chamaepitys* and *Ajuga multiflora* [11–13, 16–18]. Compound **36** was reported as a 2-*O*-glucopyranoside [20]. Phytoecdysteroids bearing a γ-lactone ring at various positions, **41**, **44** and **45**, were also isolated [21, 36]. Derivatives with a δ-lactone ring, **3**, **21**, **23**–**25**, **29**, **37**, and **46** were reported as well [14, 25, 27, 35, 36]. In addition, the reduced forms, **37** and **46** with a THP ring, and **38** and **40** with a THF ring, were isolated as acetals or hemiacetals [20, 21, 36]. Two other similar compounds with a THF ring **9** and **10**, are biosynthesized from precursors via intramolecular hydration [22]. In addition, ajugacetalsterone C (**39**) has a rare 6,8-dioxabicyclo[3.2.1]oct-2-ene structure presumably formed by an intramolecular acetalization [21]. Fujimoto and co-workers summarized biosynthesis of ecdysteroids as well as sterols in *Ajuga* hairy root in detail [39].

Withanolides (Table 2, Figure 2)

Withanolides are characteristic of Solanaceous plants, though there are rare reports on their isolation from other families. In 1999, Khan and co-workers isolated a new withanolide **47** from *Ajuga parvifora*. This is the first report of naturally occurring withanolides in Lamiaceae [40]. In subsequent studies on the chemical constituents of *A. parvifora*, the same research group obtained a series of new withanolides **48–56**, along with known **57** [41–45].

Other steroids (Table 3, Figure 3)

Compounds **61** and **67**, two C_{29} monohydroxy sterols, were isolated from *A. reptans* and their structures were elucidated by spectral methods [47]. From the aerial parts of *Ajuga salicifolia*, Akbay and co-workers isolated one new stigmastane-type sterol **72** and eight new sterol glycosides **70**, **71**, **73–78** [53, 54]. The whole plant of *Ajuga relicta* afforded two new steroids **79**, **80**, as well as two known compounds **61**, **68** [49]. A steroidal glucopyranoside **64** was isolated from *A. chamaepitys* ssp. *laevigata* [51].

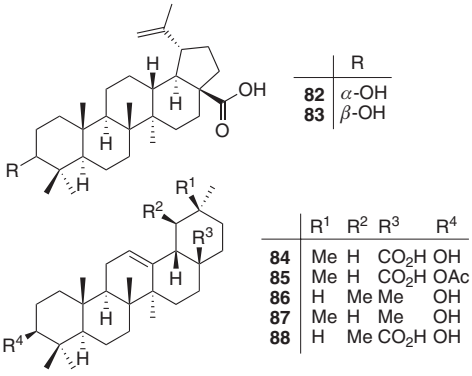


Figure 4 Triterpenoids.

Table 5 Diterpenoids.

No.	Name	Source	Part	Ref.
89	Ajugarin I	*A. remota*	leaf	[2]
		A. nipponensis	aerial part	[57]
		A. parviflora	aerial part	[58]
		A. decumbens	whole plant	[59]
90	Ajugarin II	*A. remota*	leaf	[2]
		A. parviflora	aerial part	[58]
91	Ajugarin III	*A. remota*	leaf	[2]
92	Ajugarin IV	*A. remota*	leaf	[60]
		A. ciliata var. *villosior*	aerial part	[61]
93	Ajugarin V	*A. remota*	leaf	[62]
94	Clerodin	*A. remota*	leaf	[63]
		A. bracteosa	aerial part	[64]
95	Dihydroclerodin	*A. parviflora*	aerial part	[58]
		A. bracteosa	whole plant	[46, 64]
		A. remota	aerial part	[65]
96	Ajugareptansin	*A. reptans*	aerial part	[66–68]
97	Ajugareptansone A	*A. reptans*	whole plant	[69, 70]
98	Ajugareptansone B	*A. reptans*	whole plant	[69]
99	Ivain I	*A. iva*	whole plant	[71]
100	Ivain II	*A. iva*	whole plant	[71]
		A. bracteosa	aerial part	[64]
101	Ivain III	*A. iva*	whole plant	[71]
102	Ivain IV	*A. iva*	whole plant	[71]
103	Ajugapitin (Clerodendrin D)	*A. chamaepitys*	whole plant	[72, 73]
		A. australis	aerial part	[74]
		A. decumbens	leaf	[75]
		A. remota	aerial part	[65]
		A. turkestanica	aerial part	[76]
104	14,15-Dihydroajugapitin	*A. chamaepitys*	whole plant	[72, 73]
		A. pseudoiva	leaf	[77, 78]
		A. bracteosa	whole plant	[46, 64, 79]
		A. remota	aerial part	[65]
105	Chamaepitin	*A. chamaepitys*	whole plant	[80]
		A. turkestanica	aerial part	[76]
106	Ajugamarin	*A. nipponensis*	leaf	[81–83]
		A. decumbens	whole plant	[75, 84–86]
		A. ciliata	whole plant	[87]
107	Dihydroajugamarin	*A. nipponensis*	leaf	[81]
		A. decumbens	leaf	[75]
108	Ajugamarin chlorohydrin	*A. nipponensis*	leaf	[81]
		A. ciliata	whole plant	[87]
109	2-Acetylivain I	*A. pseudoiva*	whole plant	[77]
110	Ajugamarin A2	*A. decumbens*	whole plant	[88]
		A. nipponensis	aerial part	[83]
		A. ciliata	whole plant	[87]
111	Ajugamarin B1	*A. ciliata*	whole plant	[87]
112	Ajugamarin B2	*A. nipponensis*	aerial part	[57, 83]
		A. decumbens	whole plant	[88]
113	Ajugamarin B3	*A. nipponensis*	aerial part	[57]
114	Ajugamarin B4	*A. ciliata* var. *villosior*	aerial part	[61]
115	Ajugamarin B5	*A. ciliata* var. *villosior*	aerial part	[61]
116	Ajugamarin C1	*A. nipponensis*	aerial part	[57]
		A. taiwanensis	whole plant	[89]
		A. ciliata	whole plant	[90]
117	Ajugamarin D1	*A. nipponensis*	aerial part	[57]

Table 5 (continued)

No.	Name	Source	Part	Ref.
118	Ajugamarin E1	*A. ciliata* var. *villosior*	aerial part	[61]
119	Ajugamarin E2	*A. ciliata* var. *villosior*	aerial part	[61]
120	Ajugamarin E3	*A. ciliata* var. *villosior*	aerial part	[61]
121	Ajugamarin F1	*A. ciliata* var. *villosior*	aerial part	[61]
122	Ajugamarin F2	*A. ciliata* var. *villosior*	aerial part	[61]
123	Ajugamarin F3	*A. ciliata* var. *villosior*	aerial part	[61]
124	Ajugamarin F4	*A. decumbens*	whole plant	[86, 88]
		A. parviflora	aerial part	[58]
		A. nipponensis	aerial part	[83]
125	Ajugamarin G1	*A. decumbens*	whole plant	[75, 88]
		A. ciliata	whole plant	[87]
126	Ajugamarin H1	*A. decumbens*	whole plant	[75, 88]
		A. ciliata	whole plant	[87]
127	Deacetylajugarin IV	*A. ciliata* var. *villosior*	aerial part	[61]
		A. remota	aerial part	[65]
		A. ciliata	whole plant	[91]
128	Ajugachin A	*A. chamaepitys*	aerial part	[73]
		A. reptans	aerial part	[68]
129	Ajugachin B	*A. chamaepitys*	aerial part	[73]
		A. turkestanica	aerial part	[76]
130	Ajugacumbin A	*A. decumbens*	whole plant	[59, 85, 92, 93]
		A. nipponensis	aerial part	[83]
		A. ciliata	whole plant	[87]
131	Ajugacumbin B	*A. decumbens*	whole plant	[59, 92, 93]
		A. nipponensis	aerial part	[83, 94]
		A. macrosperma	whole plant	[95]
		A. pantantha	whole plant	[95]
132	Ajugacumbin C	*A. decumbens*	whole plant	[85, 92]
133	Ajugacumbin D	*A. decumbens*	whole plant	[85, 92]
134	Ajugacumbin E	*A. decumbens*	whole plant	[84]
135	Ajugacumbin F	*A. decumbens*	whole plant	[84]
		A. ciliata	whole plant	[87]
136	Ajugacumbin G	*A. decumbens*	whole plant	[93]
137	Ajugacumbin H	*A. decumbens*	whole plant	[85]
138	Ajugacumbin J	*A. decumbens*	whole plant	[96]
139	Ajugavensin A	*A. genevensis*	aerial part	[97]
		A. reptans	aerial part	[70]
140	Ajugavensin B	*A. genevensis*	aerial part	[97]
141	Ajugavensin C	*A. genevensis*	aerial part	[97]
142	Ajugamacrin A	*A. macrosperma*	whole plant	[98]
143	Ajugamacrin B	*A. macrosperma*	whole plant	[98]
		A. taiwanensis	whole plant aerial part	[89]
		A. nipponensis	whole plant aerial part	[83]
144	Ajugamacrin C	*A. macrosperma*	whole plant	[95]
		A. pantantha	whole plant	[95]
145	Ajugamacrin D	*A. macrosperma*	whole plant	[95]
		A. pantantha	whole plant	[95]
146	Ajugamacrin E	*A. macrosperma*	whole plant	[95]
		A. pantantha	whole plant	[95]
147	Ajugapantin A	*A. macrosperma*	whole plant	[95]
		A. pantantha	whole plant	[95]
		A. taiwanensis	whole plant	[89]
		A. ciliata	whole plant	[87]
148	Deoxyajugarin-I	*A. parviflora*	aerial part	[58]
149	Ajugarin-I chlorohydrin	*A. parviflora*	aerial part	[58]

Table 5 (continued)

No.	Name	Source	Part	Ref.
150	3β-Acetoxyclerodinin C	*A. parviflora*	aerial part	[58]
151	Clerodinin A	*A. bracteosa*	whole plant	[46]
152	Clerodinin C	*A. parviflora*	aerial part	[58]
153	Clerodinin D	*A. parviflora*	aerial part	[58]
154	15-α-Ethoxy-14-hydroajugapitin	*A. parviflora*	aerial part	[58]
155	15-β-Ethoxy-14-hydroajugapitin	*A. parviflora*	aerial part	[58]
156	Lupulin A	*A. lupulina*	whole plant	[99]
		A. bracteosa	whole plant	[46]
		A. turkestanica	aerial part	[76]
		A. pseudoiva	leaf	[78]
157	Lupulin B	*A. lupulina*	whole plant	[99]
158	Lupulin C	*A. lupulina*	whole plant	[99]
159	Lupulin D	*A. lupulina*	whole plant	[99]
160	Lupulin E	*A. lupulina*	whole plant	[4]
161	Lupulin F	*A. lupulina*	whole plant	[4]
162	2β-Hydroxy-2-methylbutanoyl-3α-lupulin	*A. lupulina*	whole plant	[100]
163	6-Deacetylajugarin IV	*A. lupulina* var. *major*	whole plant	[101]
164	Ajugorientin (3β-Hydroxyajugavensin B)	*A. orientalis*	aerial part	[74]
		A. reptans	aerial part	[67, 68]
165	14,15-Dihydro-15-hydroxyajugapitin	*A. australis*	aerial part	[74]
		A. bracteosa	whole plant	[64, 79]
		A. remota	aerial part	[65]
166	Ajugatakasin A	*A. decumbens*	leaf	[75]
		A. nipponensis	aerial part	[83]
		A. ciliata	whole plant	[87]
167	Ajugatakasin B	*A. decumbens*	leaf	[75]
		A. ciliata	whole plant	[87]
168	14,15-Dehydroajugareptansin	*A. reptans*	aerial part	[67]
169	3α-Hydroxyajugamarin F4	*A. reptans*	aerial part	[67]
170	Ajugapyrin A	*A. pyramidalis*	aerial part	[102]
171	Areptin A	*A. reptans*	aerial part	[68]
172	Areptin B	*A. reptans*	aerial part	[68]
173	(15*R*)-14,15-Dihydro-15-hydroxyajugachin A	*A. laxmanii*	aerial part	[103]
174	(15*S*)-14,15-Dihydro-15-hydroxyajugachin A	*A. laxmanii*	aerial part	[103]
175	Hativene A	*A. pseudoiva*	leaf	[78]
176	Hativene B	*A. pseudoiva*	leaf	[78]
177	Hativene C	*A. pseudoiva*	leaf	[78]
178	Hativene D	*A. pseudoiva*	leaf	[104]
179	Ajugatansin A1	*A. reptans*	aerial part	[70]
180	Ajugatansin B1	*A. reptans*	aerial part	[70]
181	Ajugatansin D1	*A. reptans*	aerial part	[70]
182	Bracteonin-A	*A. bracteosa*	whole plant	[79]
183	Ajugareptone	*A. reptans*	leaf	[105]
184	Ajugalaevigatic acid	*A. chamaepitys* ssp. *laevigata*	whole plant	[51]
185	(13*S*)-15-Hydroxylabd-8(17)-en-19-oic acid (Imbricatoloic acid)	*A. chamaepitys* ssp. *laevigata*	whole plant	[51]
186	Ajugalide A	*A. taiwanensis*	whole plant	[89]
187	Ajugalide B	*A. taiwanensis*	whole plant	[89]
		A. ciliata	whole plant	[87]
188	Ajugalide C	*A. taiwanensis*	whole plant	[89]
		A. ciliata	whole plant	[87]
189	Ajugalide D	*A. taiwanensis*	whole plant	[89]
		A. ciliata	whole plant	[91]
190	Ajuganipponin A	*A. nipponensis*	aerial part	[83]
		A. ciliata	whole plant	[90]

Table 5 (continued)

No.	Name	Source	Part	Ref.
191	Ajuganipponin B	*A. nipponensis*	aerial part	[83]
		A. ciliata	whole plant	[87]
		A. decumbens	whole plant	[86]
192	14-Hydro-15-hydroxyclerodin	*A. remota*	aerial part	[65]
193	14,15-Hihydroajugachin B	*A. turkestanica*	aerial part	[76]
194	14-Hydro-15-methoxyajugachin B	*A. turkestanica*	aerial part	[76]
195	15-*epi*-Lupulin A	*A. decumbens*	whole plant	[106]
196	15-*epi*-Lupulin B	*A. bracteosa*	aerial part	[64]
197	6-*O*-Deacetylajugamarin	*A. decumbens*	whole plant	[106]
198	Ajugadecumbenins A	*A. decumbens*	whole plant	[106]
199	Ajugadecumbenins B	*A. decumbens*	whole plant	[106]
200	Ajubractin A	*A. bracteosa*	aerial part	[64]
201	Ajubractin B	*A. bracteosa*	aerial part	[64]
202	Ajubractin C	*A. bracteosa*	aerial part	[64]
203	Ajubractin D	*A. bracteosa*	aerial part	[64]
204	Ajubractin E	*A. bracteosa*	aerial part	[64]
205	3-*epi*-Caryoptin	*A. bracteosa*	aerial part	[64]
206	3-*epi*-14,15-Dihydrocaryoptin	*A. bracteosa*	aerial part	[64]
207	15-Hydroxyajubractin C	*A. bracteosa*	aerial part	[64]
208	14-Hydro-15-hydroxyajugachin A	*A. bracteosa*	aerial part	[64]
209	Ajugaciliatin A	*A. ciliata*	whole plant	[87]
210	Ajugaciliatin B	*A. ciliata*	whole plant	[87]
211	Ajugaciliatin C	*A. ciliata*	whole plant	[87]
212	Ajugaciliatin D	*A. ciliata*	whole plant	[87]
213	Ajugaciliatin E	*A. ciliata*	whole plant	[87]
214	Ajugaciliatin F	*A. ciliata*	whole plant	[87]
215	Ajugaciliatin G	*A. ciliata*	whole plant	[87]
216	Ajugaciliatin H	*A. ciliata*	whole plant	[87]
217	Ajugaciliatin I	*A. ciliata*	whole plant	[87]
218	Ajugaciliatin J	*A. ciliata*	whole plant	[87]
		A. decumbens	whole plant	[59]
219	(12*S*)-1β,6α,19-Triacetoxy-18-chloro-4α,12-dihydroxyneoclerod-13-en-15,16-olide	*A. ciliata*	whole plant	[91]
220	(12*S*,2′*S*)-12,19-Diacetoxy-18-chloro-4α,6α-dihydroxy-1β-(2-methylbutanoyloxy)neoclerod-13-en-15,16-olide	*A. ciliata*	whole plant	[91]
221	(12*S*)-6α,18,19-Triacetoxy-4α,12-dihydroxy-1β-tigloyloxyneoclerod-13-en-15,16-olide	*A. ciliata*	whole plant	[91]
222	(12*S*)-6α-Acetoxy-4α,18-epoxy-12-hydroxy-19-tigloyloxyneoclerod-13-en-15,16-olide	*A. ciliata*	whole plant	[90]
223	6α,18-Diacetoxy-4α-hydroxy-19-tigloyloxyneoclerod-13-en-15,16-olide	*A. ciliata*	whole plant	[90]
224	Ajugamarin A2 chlorohydrin	*A. ciliata*	whole plant	[87]
225	6α,19-Diacetoxy-4α-hydroxy-1β-tigloyloxyneoclerod-12-en-15-oic acid methyl ester-16-aldehyde	*A. decumbens*	whole plant	[59]
226	(12*S*)-18,19-Diacetoxy-4α,6α,12-trihydroxy-1β-tigloyloxyneoclerod-13-en-15,16-olide	*A. decumbens*	whole plant	[59]
227	4α,6α-Dihydroxy-18-(4′-methoxy-4′-oxobutyryloxy)-19-tigloyloxyneoclerod-13-en-15,16-olide	*A. decumbens*	whole plant	[59]
228	(12*S*)-1α,19-Epoxy-6α,18-diacetoxy-4α,12-dihydroxyneoclerod-13-en-15,16-olide	*A. decumbens*	whole plant	[86]
229	(12*S*)-6α,19-Diacetoxy-18-chloro-4α-hydroxy-12-tigloyloxyneoclerod-13-en-15,16-olide	*A. decumbens*	whole plant	[86]
230	(12*S*,2″*S*)-6α,19-Diacetoxy-18-chloro-4α-hydroxy-12-(2-methylbutanoyloxy)neoclerod-13-en-15,16-olide	*A. decumbens*	whole plant	[86]
231	Ajugacumbin A chlorohydrin	*A. ciliata*	whole plant	[87]
		A. decumbens	whole plant	[59]
232	Ajuforrestin A	*A. forrestii*	whole plant	[107]
		A. decumbens	aerial part	[7]

Table 5 (continued)

No.	Name	Source	Part	Ref.
233	Ajuforrestin B	*A. forrestii*	whole plant	[107]
		A. decumbens	aerial part	[7]
234	Ajugaside A	*A. decumbens*	whole plant	[108]
235	Ajudecumin A	*A. decumbens*	aerial part	[7]
236	Ajudecumin B	*A. decumbens*	aerial part	[7]
237	Ajudecumin C	*A. decumbens*	aerial part	[7]
238	Ajudecumin D	*A. decumbens*	aerial part	[7]
239	Carnosol	*A. forrestii*	whole plant	[109]
240	Epiisorosmanol	*A. forrestii*	whole plant	[109]
241	2,11,12-Trihydroxy-7,20-epoxy-8,11,13-abietatriene	*A. forrestii*	whole plant	[109]
242	Epirosmanol	*A. forrestii*	whole plant	[109]
243	7-Methoxyrosmanol	*A. forrestii*	whole plant	[109]
244	7-Ethoxyrosmanol	*A. forrestii*	whole plant	[109]
245	2α,3β,11,12-Tetrahydroxy-7β,20-epoxy-8,11,13-abietatriene	*A. forrestii*	whole plant	[109]

Triterpenoids (Table 4, Figure 4)

In 1997, two lupan triterpenoids **82** and **83** were isolated from the aerial parts of *Ajuga macrosperma* [56]. Oleananes **84** and **85** were two known triterpenoids isolated from *A. relicta* [49]. From *A. chamaepitys* ssp. *laevigata*, two ursanes and one oleanane **86–88** were isolated [51].

Diterpenoids (Table 5, Figure 5)

Ajuga species are rich in diterpenoids. With respect to the carbocyclic skeleton, *Ajuga* diterpenoids roughly belong to two groups: neoclerodane and abietane types.

Neoclerodanes

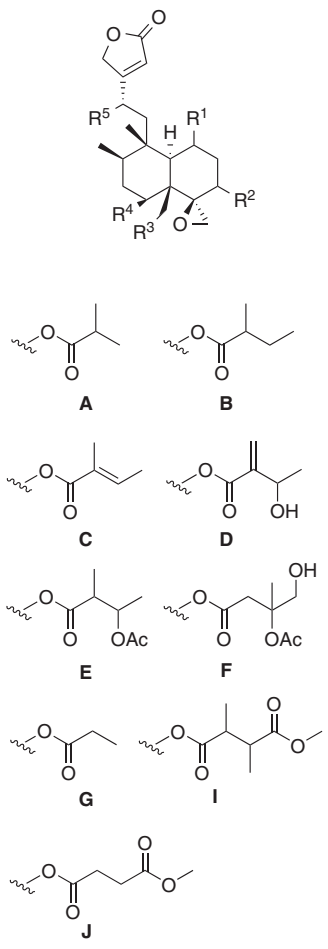
Most of the neoclerodane diterpenoids produced by species of the genus *Ajuga* contain a substituted decalin with a 4α,18-oxirane ring and two oxygenated substituents bound to C(6) and C(19) [110]. The side chain features several moieties with the most common being: (i) a butenolide function (α-substituted α,β-unsaturated γ-lactone, or 13-en-15,16-olide) as in **89–93** isolated from *A. remota* [2, 60, 62]; (ii) a tetrahydrofurofuran as in **94** reported as a constituent of *A. remota* [63]; and (iii) a hexahydrofurofuran as in **95** reported as a component of *Ajuga parviflora* [58]. In 1983, three new bitter principles, **106–108**, were isolated from the leaves of *A. nipponensis*. The β-hydrin structure of **108** was confirmed by treatment of **106** with methanolic HCl [81, 82]. In addition, by reinvestigation of the aerial parts of *A. nipponensis*, four new bitter neoclerodanes **112**, **113**, **116**, **117** and a known diterpenoid **89** were isolated [57]. The configuration of methoxy group at C(15) (R^5) of lupulin A (**156**) [99] was revised to be α by Huang and co-workers [106]. Consequently, hativene D (**178**) [104] is not 15-*epi*-lupulin (**195**) but lupulin (**156**). The structure of 15-*epi*-lupulin B (**196**) [64] with a 2β-OH group coincides with lupulin A **156**.

Abietanes

A new abietane diglucopyranoside **234** was isolated from the whole plants of *A. decumbens* [108]. In the course of the search for bioactive metabolites with anticancer effects, Wang et al. isolated four new rearranged abietane hydroquinones **235–238**, together with two known abietanes **232** and **233** from the aerial parts of *A. decumbens* collected in China [7].

Sesquiterpenoids (Table 6, Figure 6)

Only a few other sesquiterpenoids, bisabolene, eudesmanolides and seco-sesquiterpenoids, were reported. Compound **246**, a bisabolene sesquiterpenoid, was isolated from the aerial parts of *A. decumbens* [7]. Research on *Ajuga forrestii* resulted in the isolation of four eudesmane sesquiterpene lactones **247–250**. Among them, compounds **247–249** are new. Compound **249** exhibits weak cytotoxic activity against HepG2 and MCF-7 human cell lines [109]. Four megastigmane derivatives **251–254** and **257** were isolated in 2012 [7, 112]. A new ionone glycoside **255** was also isolated from this plant. This is the



	R^1	R^2	R^3	R^4	R^5
89	H	H	OAc	OAc	H
90	H	H	OAc	OH	H
93	H	H	H	OAc	H
97	=O	α-**B**	OAc	OAc	H
106	α-**C**	H	OAc	OAc	OH
107	α-**B**	H	OAc	OAc	OH
110	α-**C**	H	OAc	OAc	OAc
111	α-**B**	H	OAc	OAc	OH
112	α-**B**	H	OAc	OAc	OAc
113	α-**B**	H	OAc	OH	OH
114	α-**B**	H	OH	OAc	OH
115	α-**B**	H	OH	OAc	OAc
116	α-OH	H	OAc	OAc	OH
118	α-OH	H	OH	OAc	**B**
119	α-OH	H	OAc	OH	**B**
120	α-OH	H	OAc	OAc	**B**
121	H	H	OH	OH	**B**
122	H	H	OH	OAc	**B**
123	H	H	OAc	OH	**B**
124	H	H	OAc	OAc	**B**
125	α-**C**	H	OAc	OAc	**B**
126	α-**B**	H	OAc	OAc	**C**
130	H	H	**C**	OAc	H
131	H	H	**C**	OH	H
132	β-OAc	α-OAc	**C**	OAc	H
133	H	α-OH	**C**	OAc	H
134	β-OAc	α-OAc	**D**	OAc	H
136	H	H	**D**	OAc	H
137	β-OAc	α-OAc	**C**	OAc	OAc
142	α-OAc	H	OAc	OAc	**A**
143	α-OAc	H	OAc	OAc	**B**
144	α-**A**	H	OAc	OAc	**A**
145	α-**A**	H	OAc	OAc	**B**
146	α-**B**	H	OAc	OAc	**A**
147	α-OAc	H	OAc	OAc	OAc
166	α-**C**	H	OAc	OAc	**C**
167	α-**B**	H	OAc	OAc	**B**
169	H	β-OH	OAc	OAc	**B**
179	α-OH	α-**B**	OAc	OAc	H
180	H	α-OH	OAc	OAc	**B**
183	=O	α-OH	OAc	OAc	**B**
186	α-OH	α-OH	OAc	OAc	OAc
187	α-OAc	H	OAc	OAc	OH
188	H	H	OAc	OAc	OH
190	α-OAc	H	OAc	OAc	**C**
191	H	H	OAc	OAc	**C**
197	α-**C**	H	OAc	OH	OH
198	H	H	OAc	OH	OAc
199	H	H	OAc	OH	**C**
222	H	H	**C**	OAc	OH

Figure 5 Diterpenoids.

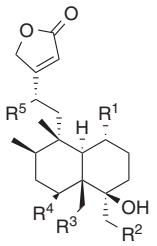
first report of the occurrence of ionone glycosides in *Ajuga* species [111].

Monoterpenoids (Table 7, Figure 7)

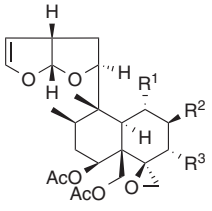
Major monoquiterpenoids isolated from the genus *Ajuga* belong to iridoids. In 1974, Guiso and co-workers isolated three iridoid glucopyranosides **258–260** from *A. reptans*. Compound **259** is an 8-*O*-acetyl derivative of **260** and a 6-epimer of 8-*O*-acetylmioporoside **271** [113, 117]. Four new iridoid glucopyranoside *cis*- and *trans*-*p*-coumaroyl esters **263–266** were isolated from methanol extract of the dried plant of *A. decumbens*, together with the known compounds **258** and **262** [116]. Compound **270**, isolated from the leaves of *Ajuga pseudoiva*, possesses an unusual 13-membered macrocyclic structure [121].

Flavonoids (Table 8, Figure 8)

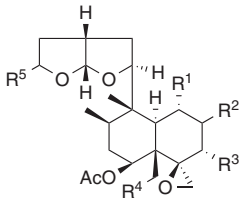
Flavonoids **288–305** including flavones and flavonols isolated from the genus *Ajuga* are outlined in Table 8.



	R^1	R^2	R^3	R^4	R^5
91	H	OH	OAc	OAc	H
108	**C**	Cl	OAc	OAc	OH
117	**B**	OAc	OAc	OH	OH
135	H	OH	**C**	OH	H
149	H	Cl	OAc	OAc	H
209	**B**	Cl	OAc	OAc	**B**
210	OAc	Cl	OAc	OAc	**B**
211	**C**	Cl	OAc	OAc	**B**
212	**B**	Cl	OAc	OAc	OAc
213	**C**	Cl	OAc	OAc	**C**
217	H	OAc	**C**	OH	H
218	H	**C**	OH	OH	H
219	OAc	Cl	OAc	OAc	OH
220	**B**	Cl	OAc	OH	OAc
221	**C**	OAc	OAc	OAc	OH
223	H	OAc	**C**	OAc	H
224	**C**	Cl	OAc	OAc	OAc
226	**C**	OAc	OAc	OH	OH
227	H	**J**	**C**	OH	H
229	H	Cl	OAc	OAc	**C**
230	H	Cl	OAc	OAc	**B**
231	H	Cl	**C**	OAc	H



	R^1	R^2	R^3
94	H	H	H
103	H	OH	**B**
128	H	OH	**A**
129	H	OH	**I**
160	H	OH	**G**
168	**B**	H	OH
172	**C**	H	OH
200	H	H	**B**
201	H	H	**A**
205	H	H	OAc



	R^1	R^2	R^3	R^4	R^5
95	H	H	H	OAc	H
96	**B**	H	OH	OAc	H
99	H	α-OH	**A**	OAc	H
100	H	H	**A**	OAc	H
101	H	α-OH	**A**	OAc	OEt
102	H	α-OH	**B**	OAc	H
104	H	β-OH	**B**	OAc	H
105	H	β-OH	**E**	OAc	OH
109	H	α-OAc	**A**	OAc	H
139	**B**	H	H	OAc	H
140	**C**	H	H	OAc	H
141	OH	H	H	**C**	H
150	H	H	Ac	OAc	β-OEt
151	H	H	H	OAc	β-OMe
152	H	H	H	OAc	β-OEt
153	H	H	H	OAc	α-OEt
154	H	β-OH	**B**	OAc	α-OEt
155	H	β-OH	**B**	OAc	β-OEt
156	H	β-OH	**B**	OAc	α-OMe
157	H	H	**B**	OAc	α-OMe
159	H	H	H	OAc	α-OMe
161	H	β-OH	**G**	OAc	H
162	H	β-OH	**B**	OAc	H
164	**C**	H	OH	OAc	H
165	H	β-OH	**B**	OAc	OH
171	OH	β-OAc	**B**	OAc	H
173	H	β-OH	**A**	OAc	β-OH
174	H	β-OH	**A**	OAc	α-OH
175	H	β-OH	**A**	OAc	β-OMe
176	H	β-OH	**A**	OAc	α-OMe
177	H	α-OH	**A**	OAc	α-OMe
178	H	β-OH	**B**	OAc	α-OMe
181	**C**	α-OH	**B**	OAc	α-OMe
182	H	H	H	**F**	OMe
192	H	H	H	OAc	OH
193	H	β-OH	**E**	OAc	H
194	H	β-OH	**E**	OAc	OMe
195	H	β-OH	**B**	OAc	β-OMe
196	H	β-OH	**B**	OAc	α-OMe
202	H	H	**B**	OAc	H
203	H	β-OH	**A**	OAc	H
204	H	H	OH	OAc	H
206	H	H	OAc	OAc	H
207	H	H	**B**	OAc	OH
208	H	H	**A**	OAc	OH

Figure 5 (continued)

Polyketides and alkaloids (Table 9, Figure 9)

From the leaves of *A. iva*, three new homologous 1,3-diglycerides **331–333** and compound **339** were obtained. In 1986, Takasaki and co-workers isolated three phenethyl alcohol glycosides **323–325**, including a new derivative **323** [108]. A phenylalanine derivative **346** was isolated from this plant recently [96]. In addition, Yu and co-workers isolated a phthalic ester **321** from the aerial parts of *A. multiflora* [18].

During the search for bioactive metabolites from *A. pseudoiva* leaves, Ben and co-workers isolated five novel monoglycerides **334–338**, two novel cinnamic acids **318**, **319** and one new steroid **63**, along with five known compounds **61**, **62**, **331–333**. Three compounds **331–333** show significant antifeedant activity, which might be associated with the presence of two β-hydroxyalkanoic moieties in each compound [38, 48, 50, 132]. A phytochemical investigation on *A. parviflora* resulted in the isolation of quinols **306–312** and pyrrolizidine alkaloids **343–345**. Derivatives **306–308** are new compounds. Compound **309**, isolated previously from the leaves and branches of *Jacaranda* species, display cytotoxic and antitumor activities. Three pyrrolizidine alkaloids **343–345** were reported for the first time from this plant [44, 45, 128, 129].

The plant of *Ajuga bracteosa* afforded several new compounds including unsaturated ketone **341**, phthalic

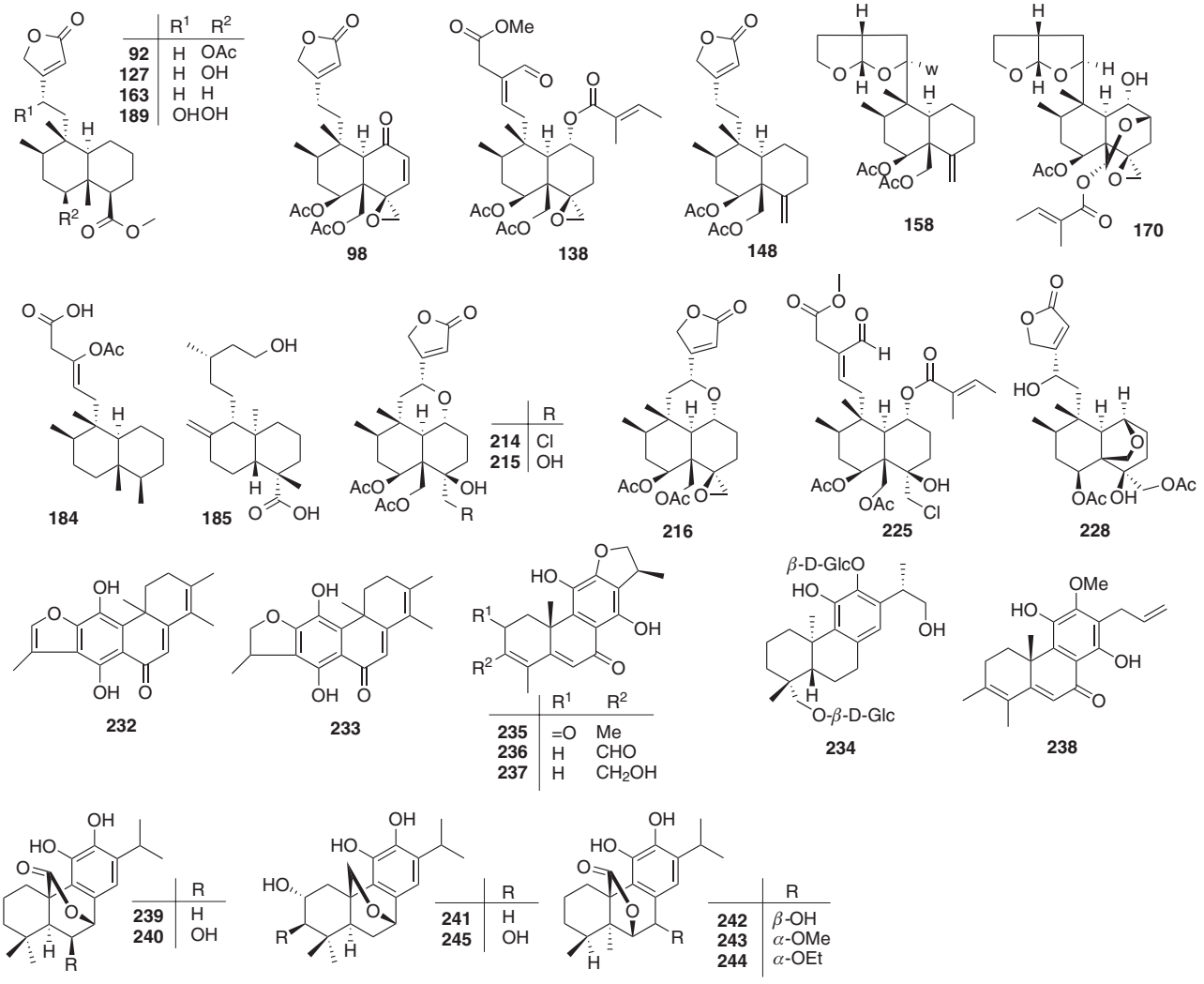


Figure 5 (continued)

Table 6 Sesquiterpenoids.

No.	Name	Source	Part	Ref.
246	Glecholone	*A. decumbens*	aerial part	[7]
247	3α-Acetoxy-1α,8β-dihydroxyeudesm-7(11)-en-8,12-olide	*A. forrestii*	whole plant	[109]
248	3α-Acetoxy-1α-hydroxyeudesm-8,7(11)-dien-8,12-olide	*A. forrestii*	whole plant	[109]
249	1α-Acetoxy-8α-oxyethyl-2-oxoeudesman-3,7(11)-dien-8,12-olide	*A. forrestii*	whole plant	[109]
250	1α-Acetoxy-8α-hydroxy-2-oxoeudesman-3,7(11)-dien-8,12-olide	*A. forrestii*	whole plant	[109]
251	(6*R*,7*E*,9*R*)-9-Hydroxy-4,7-megastigmadien-3-one	*A. decumbens*	aerial part	[7]
252	(3*S*,5*R*,6*S*,7*E*)-5,6-Epoxy-3-hydroxy-7-megastigmen-9-one	*A. decumbens*	aerial part	[7]
253	(6*E*,9*S*)-9-Hydroxy-4,6-megastigmadien-3-one	*A. decumbens*	aerial part	[7]
254	6-Hydroxy-4,7-megastigmadiene-3,9-dione	*A. decumbens*	aerial part	[7]
255	4β-Hydroxy-7,8-dihydro-3-oxo-β-ionol 9-*O*-β-D-glucopyranoside	*A. salicifolia*	aerial part	[111]
256	Corchoionoside C	*A. salicifolia*	aerial part	[111]
257	Loliolide	*A. decumbens*	whole plant	[112]

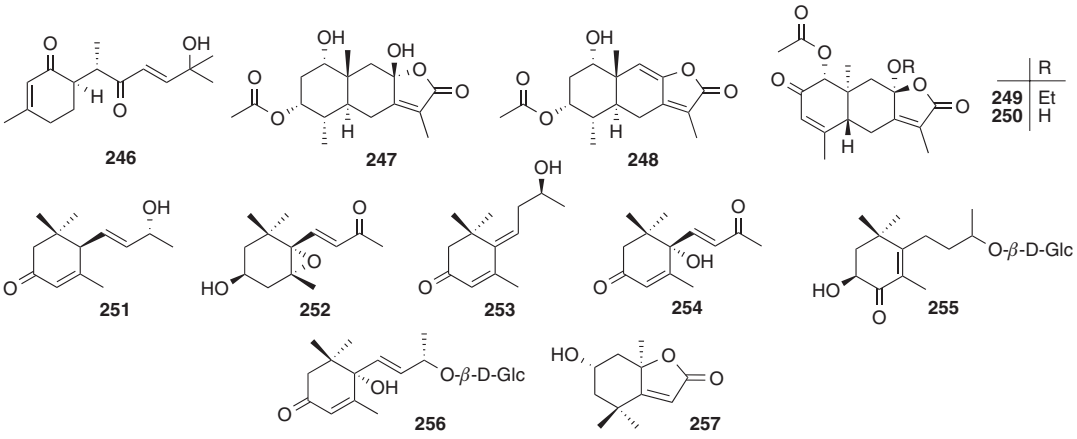


Figure 6 Sesquiterpenoids.

Table 7 Iridoids.

No.	Name	Source	Part	Ref.
258	Reposide	*A. reptans*	whole plant	[113–115]
		A. decumbens	whole plant	[116]
259	Ajugoside	*A. reptans*	whole plant	[117]
260	Ajugol	*A. reptans*	whole plant	[117]
261	Jaranidoside	*A. spectabilis*	whole plant	[118]
262	8-*O*-Acetylharpagide	*A. multiflora*	whole plant	[119]
		A. remota	aerial part	[6]
		A. decumbens	whole plant	[116]
		A. iva	aerial part	[120]
		A. reptans	whole plant	[114, 115]
263	Decumbeside A	*A. decumbens*	whole plant	[116]
264	Decumbeside B	*A. decumbens*	whole plant	[116]
265	Decumbeside C	*A. decumbens*	whole plant	[116]
266	Decumbeside D	*A. decumbens*	whole plant	[116]
267	Harpagide	*A. iva*	aerial part	[120]
		A. reptans	whole plant	[114, 115]
268	6-Deoxyharpagide	*A. iva*	aerial part	[120]
269	Ajureptoside	*A. reptans*	whole plant	[114]
270	7-*O*-6′-*O*-Malonylcachinesidic acid	*A. pseudoiva*	leaf	[121]
271	8-*O*-Acetylmioporoside	*A. salicifolia*	aerial part	[111]
272	Galiridoside	*A. taiwanensis*	whole plant	[19]
273	Teuhircoside	*A. taiwanensis*	whole plant	[19]
274	6-Keto-8-acetylharpagide	*A. remota*	aerial part	[122]
275	6,7-Dehydro-8-acetylharpagide	*A. remota*	aerial part	[122]
276	7,8-Dehydroharpagide	*A. remota*	aerial part	[122]
277	8-Acetylharpagide 6-*O*-β-glucopyranoside	*A. remota*	aerial part	[122]
278	Harpagide 6-*O*-β-glucopyranoside	*A. remota*	aerial part	[122]
279	2′,3′-Diacetylharpagide	*A. remota*	underground part	[123]
280	6′-*O*-Rhamnosylharpagide	*A. remota*	underground part	[123]
281	6′-*O*-Galloyl-7,8-dehydroharpagide	*A. remota*	underground part	[123]
282	6-*O*-Xylosylharpagoside-B	*A. remota*	underground part	[123]
283	Ajureptaside A	*A. reptans*	whole plant	[115]
284	Ajureptaside B	*A. reptans*	whole plant	[115]
285	Ajureptaside C	*A. reptans*	whole plant	[115]
286	Ajureptaside D	*A. reptans*	whole plant	[115]
287	6-*epi*-8-*O*-Acetylharpagide	*A. reptans*	whole plant	[115]

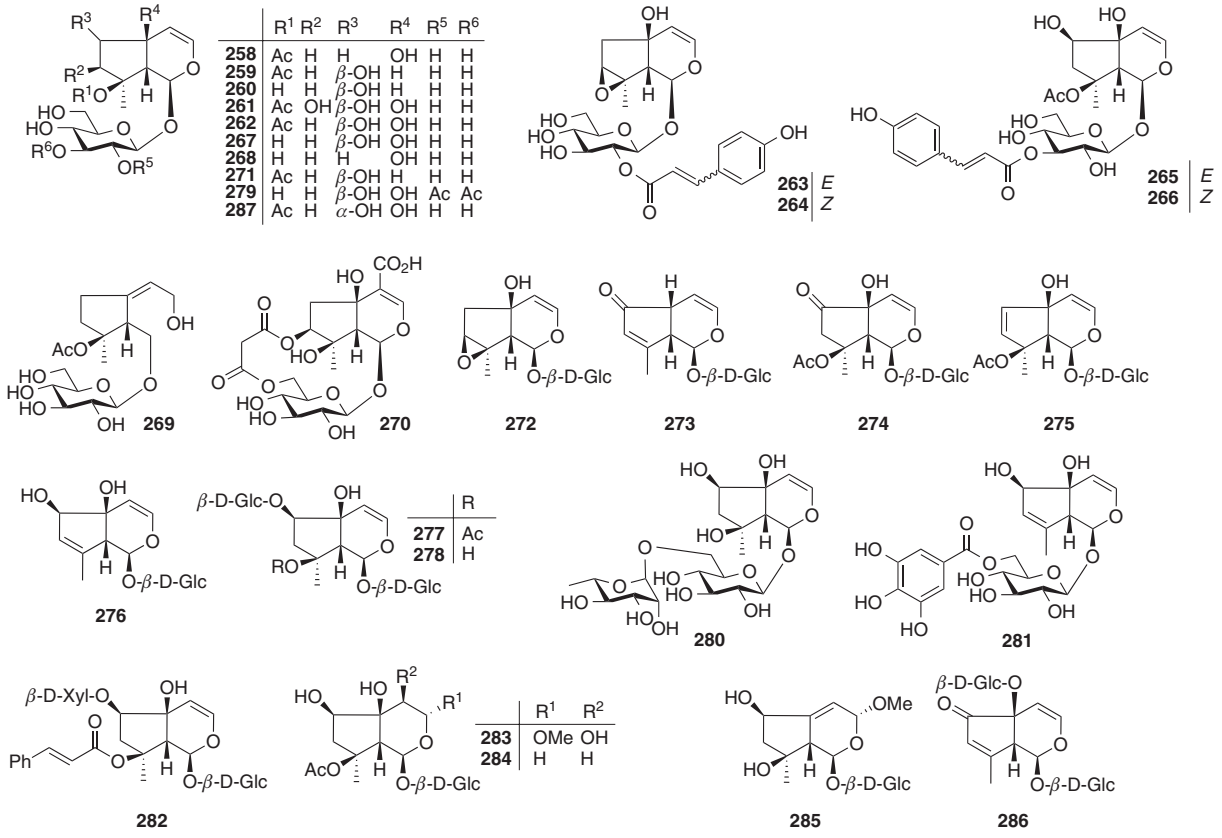
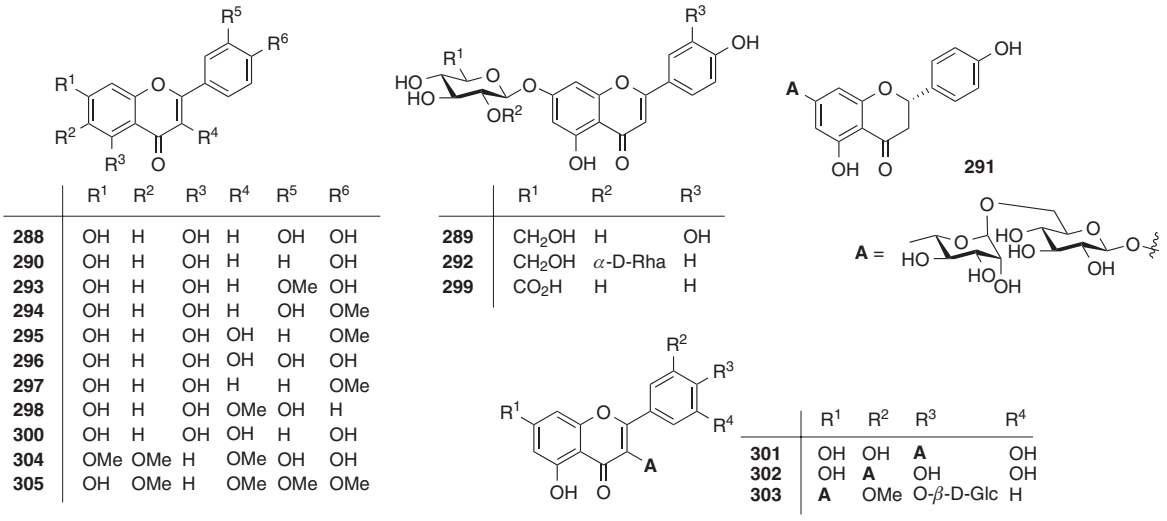


Figure 7 Iridoids.

Table 8 Flavonoids.

No.	Name	Source	Part	Ref.
288	Luteolin	*A. chia*	aerial part	[124]
		A. lupulin	whole plant	[101, 125]
289	Luteolin 7-*O*-glucopyranoside	*A. chia*	aerial part	[124]
		A. lupulina	whole plant	[101, 125]
290	Apigenin	*A. chia*	aerial part	[124]
		A. multiflora	aerial part	[126]
		A. forrestii	whole plant	[107]
291	Naringin	*A. iva*	aerial part	[15]
292	Apigenin 7-*O*-neohesperidoside	*A. iva*	aerial part	[15]
293	Chrysoriol	*A. lupulina*	whole plant	[125]
294	Diosmetin	*A. lupulina*	whole plant	[125]
295	Kaempferide	*A. lupulina*	whole plant	[125]
296	Quercetin	*A. lupulina*	whole plant	[125]
297	Acacetin	*A. forrestii*	whole plant	[107]
298	Gnetifolin B	*A. forrestii*	whole plant	[107]
299	Apigenin 7-glucuronide	*A. multiflora*	aerial part	[18]
300	Kaempferol	*A. taiwanensis*	whole plant	[19]
301	Myricetin 3-*O*-rutinoside 4′-*O*-rutinoside	*A. remota*	aerial part	[122]
302	Myricetin 3-*O*-rutinoside 3′-*O*-rutinoside	*A. remota*	aerial part	[122]
303	Isorhamnetin 3-*O*-rutinoside 7-*O*-rutinoside 4′-*O*-β-glucopyranoside	*A. remota*	aerial part	[122]
304	3,4′-Dihydroxy-3,6,7-trimethoxyflavone	*A. bracteosa*	whole plant	[127]
305	7-Hydroxy-3,6,3′,4′-tetramethoxyflavone	*A. bracteosa*	whole plant	[127]



	R^1	R^2	R^3	R^4	R^5	R^6
288	OH	H	OH	H	OH	OH
290	OH	H	OH	H	H	OH
293	OH	H	OH	H	OMe	OH
294	OH	H	OH	H	OH	OMe
295	OH	H	OH	OH	H	OMe
296	OH	H	OH	OH	OH	OH
297	OH	H	OH	H	H	OMe
298	OH	H	OH	OMe	OH	H
300	OH	H	OH	OH	H	OH
304	OMe	OMe	H	OMe	OH	OH
305	OH	OMe	H	OMe	OMe	OMe

	R^1	R^2	R^3
289	CH_2OH	H	OH
292	CH_2OH	α-D-Rha	H
299	CO_2H	H	H

	R^1	R^2	R^3	R^4
301	OH	OH	**A**	OH
302	OH	**A**	OH	OH
303	**A**	OMe	O-β-D-Glc	H

Figure 8 Flavonoids.

ester **322**, phenolic compound **320**, two sphingolipids **352**, **353** and a long-chain polyhydroxy acid **342** [10, 130, 134].

Biological activity

Antifeedant and larvicidal activity

A neoclerodane **103** was isolated from the leaves of *A. decumbens* as a feeding stimulant for *Athalia rosae ruficornis* [75]. Three new neoclerodanes **164**, **168**, **169** were isolated from the aerial parts of *A. reptans* cv. catlins giant. Insect antifeedant testing revealed that **168** has significant activity against sixth stadium larvae of *Spodoptera littoralis* [67]. A series of active clerodanes **104**, **156**, **175–177** were isolated from the acetone extract of *A. pseudoiva* leaves by bioassay-guided chromatography. The behavioral responses of *Spodoptera littoralis* larvae to all clerodanes showed strong antifeedant activity at 100 to 1 mg/L. In addition, this study also indicated that a methoxy group at C(15), either in the α- or β-position, might decrease antifeedant activity [9]. Manguro and co-workers tested larvicidal activity of the extracts of *A. remota* using second instar *Aedes aegypti* larvae [123]. The ethyl acetate extract is toxic with LC_{50} value of 5.30 µg/L, while the methanol extract displays weak toxicity with an LC_{50} of 65.94 µg/L. Compound **81**, obtained from the ethyl acetate extract, is the active component with an LC_{50} value of 4.40 µg/L.

Antimicrobial activity

Compounds **156–161** are six new neoclerodanes isolated from *Ajuga lupulina*. The diterpenoids **156** and **160** show strong activity against *Pseudomonas aeruginosa* and *Escherichia coli* (inhibitory zones are 3–5 mm and 3.5–4.5 mm, respectively, at a concentration of 0.02 mg/mL). In addition, **156** displays weak activity against *Staphylococcus aureus* (1.5 mm). The antibacterial activity of **161** against *P. aeruginosa* (2.1 mm) and *E. coli* (2.0 mm) is poor compared to **156** and **160**. Compound **157** exhibits weak antibacterial activity against *S. aureus* and *E. coli* (1.2 mm) [4, 99]. In 2001, Kariba tested the extracts of *A. remota* for *in vitro* antifungal activity. The petroleum ether and methanol extracts exhibit antifungal activity against the dermatophytic fungi *Trichophyton mentagrophytes* and *Microsporum gypseum* [5]. Ergosterol 5,8-endoperoxide **81**, isolated from the methanol extract of *A. remota*, shows activity against *Mycobacterium tuberculosis* [55].

Antimalarial activity

Ajuga remota is commonly used as medicinal herb for malaria treatment in Kenya. Three isolates, **81**, **89** and **262**, were tested for their *in vitro* antiplasmodial activity. Compound **89** is moderately active against a chloroquine-sensitive (FCA 20/GHA) strain of *Plasmodium falciparum*, with an IC_{50} of 23.0 µM, compared to a 0.041 µM IC_{50} for chloroquine. Compared to **89**, compound **262** is approximately 3 times as potent. Compound **262** is also equally potent towards chloroquine-sensitive (FCA 20/GHA) and

Table 9 Polyketides and alkaloids.

No.	Name	Source	Part	Ref.
306	Ethyl (1-acetoxy-4-oxo-2,5-cyclohexadien-1-yl)acetate	*A. parviflora*	whole plant	[128]
307	Methyl (1-acetoxy-4-oxo-2,5-cyclohexadien-1-yl)acetate	*A. parviflora*	whole plant	[128]
308	Ethyl (1-hydroxy-4-oxo-2,5-cyclohexadien-1-yl)acetate	*A. parviflora*	whole plant	[128]
309	Methyl (1-hydroxy-4-oxo-2,5-cyclohexadien-1-yl)acetate	*A. parviflora*	whole plant	[128]
310	(1-Hydroxy-4-oxo-2,5-cyclohexadien-1-yl)acetic acid	*A. parviflora*	whole plant	[129]
311	2-Hydroxy-4β-methyl-4α-(β-D-glucopyranoside)-2,5-cyclohexadien-1-one	*A. parviflora*	whole plant	[129]
312	Methyl 2-(2,2-dimethyl-6-oxo-7-dihydro-1,3-benzodioxol-3(6*H*)-yl)acetate	*A. parviflora*	whole plant	[129]
313	6,7-Dihydroxycoumarin (Esculetin)	*A. decumbens*	whole plant	[52]
314	Coumarin	*A. laxmanii*	aerial part	[103]
315	Vanillic acid	*A. decumbens*	whole plant	[112]
		A. taiwanensis	whole plant	[19]
316	Melilotic acid methyl ester	*A. laxmanii*	aerial part	[103]
317	Methyl caffeate	*A. decumbens*	whole plant	[112]
318	Methyl (*E*)-4-acetoxy-3-methoxycinnamate	*A. pseudoiva*	leaf	[38]
319	Methyl (*E*)-4-acetoxycinnamate	*A. pseudoiva*	leaf	[38]
320	Ajuganane	*A. bracteosa*	whole plant	[127]
321	Bis(2-ethylhexyl) phthalate	*A. multiflora*	aerial part	[18]
322	Bis(2*S*-methylheptyl) phthalate	*A. bracteosa*	whole plant	[130]
323	Galactosylmartynoside	*A. decumbens*	whole plant	[108]
324	Martynoside	*A. decumbens*	whole plant	[108]
325	Darendoside B	*A. decumbens*	whole plant	[108]
326	Lavandulifolioside	*A. salicifolia*	aerial part	[111]
327	Leonoside A	*A. salicifolia*	aerial part	[111]
328	Leonoside B	*A. salicifolia*	aerial part	[111]
329	1-Ethenylhexyl 6-*O*-β-L-arabinopyranosyl-2-*O*-β-D–glucopyranosyl-β-D-glucopyranoside	*A. decumbens*	whole plant	[52]
330	Butyl β-D-fructopyranoside	*A. decumbens*	whole plant	[52]
331	Ivade A	*A. iva*	leaf	[131]
		A. pseudoiva	leaf	[48]
332	Ivade B	*A. iva*	leaf	[131]
		A. pseudoiva	leaf	[48]
333	Ivade C	*A. iva*	leaf	[131]
		A. pseudoiva	leaf	[48]
334	Hizivaide A	*A. pseudoiva*	leaf	[132]
335	Hizivaide B	*A. pseudoiva*	leaf	[132]
336	Hizivaide C	*A. pseudoiva*	leaf	[132]
337	Hizivaide D	*A. pseudoiva*	leaf	[132]
338	Hizivaide E	*A. pseudoiva*	leaf	[132]
339	Methyl 3-hydroxyhexadecanoate	*A. iva*	leaf	[133]
340	(10*E*,15*Z*)-9,12,13-Trihydroxyoctadeca-10,15-dienoic acid	*A. decumbens*	whole plant	[112]
341	Heptacos-3-en-25-one	*A. bracteosa*	aerial part	[134]
342	Bractic acid	*A. bracteosa*	whole plant	[10]
343	Ligularinine	*A. parviflora*	whole plant	[44]
344	Senecionine	*A. parviflora*	whole plant	[45]
345	Integerrimine	*A. parviflora*	whole plant	[45]
346	Aurantiamide acetate	*A. decumbens*	whole plant	[96]
347	Pheophytin-a	*A. taiwanensis*	whole plant	[19]
348	Pheophytin-b	*A. taiwanensis*	whole plant	[19]
349	13^2-Hydroxy(13^2-*S*)pheophytin-a	*A. taiwanensis*	whole plant	[19]
351	Nicotinic acid	*A. taiwanensis*	whole plant	[19]
352	Bractin A	*A. bracteosa*	whole plant	[10]
353	Bractin B	*A. bracteosa*	whole plant	[10]

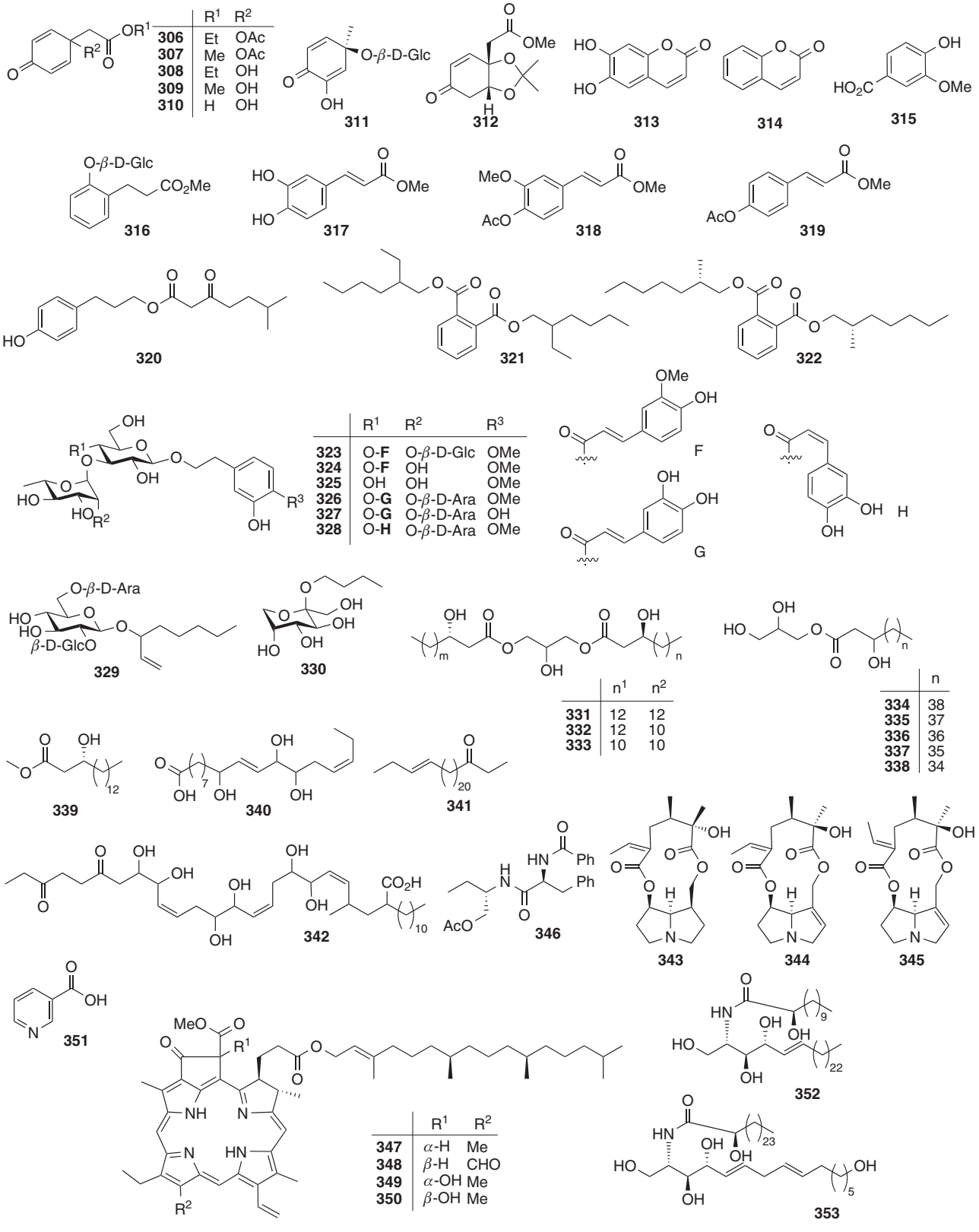


Figure 9 Polyketides and alkaloids.

chloroquine-resistant (W2) strains [6]. An excellent review article summarizes antimalarial activity of compounds contained in *A. remota* and *A. bracteosa* [135].

Anti-inflammatory activity

Gautam et al. tested a 70% ethanol extract of *A. bracteosa* whole plants in a mice acute inflammation model based on topical application of TPA. The result showed that the extract exhibits a remarkable and dose-dependent anti-inflammatory activity at 0.5 and 1.0 mg/ear. In addition, it showed a significant *in vitro* COX-1 and COX-2 inhibitory activity at 25 and 50 μg/mL. Among the isolates from the bioactive extract, compound **156** exhibited the highest inhibition of COX-1, and compound **268** displayed the highest inhibition of COX-2 [136]. The compounds **342**, **352**, **353** exhibited remarkable inhibition of lipoxygenase. Compound **342** was more active than baicalein (IC_{50} = 22.4 μM) with an IC_{50} of 10.0 μM [10].

Hypoglycemic activity

Ajuga iva has been used as traditional medicine to control diabetes mellitus for many centuries. In 2002, a study to examine the hypoglycemic effect of *A. iva* was carried out, and the results demonstrated that *A. iva* aqueous extract exhibits strong hypoglycemic activity. Lyophilized aqueous extract of *A. iva* whole plant was found to decrease plasma glucopyranose levels of streptozotocin-induced diabetic rats from 337 to 102.2 mg/dL after 6 h of oral administration. Furthermore, repeated oral administration significantly reduced plasma glucopyranose levels after 1 week of treatment (112 mg/dL at 1 week vs. 337 mg/dL at the baseline values) [137].

Cytotoxic activity

Compounds **70–75** are five new sterol glycosides isolated from a methanol extract of the aerial parts of *A. salicifolia*. Their cytotoxicity against HeLa cells (KB), human T cell leukemia (Jurkat), and peripheral mononuclear blood cells (PMBC) have been evaluated. Compounds **70–74** significantly inhibit the viability and growth of Jurkat T cells at concentrations below 10 μM. Compound **73** is the most active substance with an IC_{50} values of 3 μM, followed by **70** (IC_{50} = 6 μM). An additional glucopyranose substituent leads to weaker cytotoxicity against Jurkat T cells, as observed for **71** (IC_{50} = 10 μM) and **74** (IC_{50} = 8 μM). Compound **70** induces cell-cell contacts in a Jurkat T cell population, and remarkably up-regulated mRNA levels of the cell-cycle regulator cyclin D1, which might be an indication for cell differentiation [53]. In 2003, Akbay and co-workers investigated the cytotoxicity of sterols obtained from *A. salicifolia* against KB (HeLa) and Jurkat T cancer cells. This study demonstrated that compound **72** is active against KB cells with an IC_{50} of 1 μg/mL, while the corresponding 3-*O*-*β*-glucopyranoside, compound **76**, is less potent (IC_{50} = 13 μg/mL) [54]. Four new *A. decumbens* abietane diterpenoids, **235–238**, were evaluated for *in vitro* inhibition of cell proliferation. The diterpenoids **235** and **237** exhibit moderate cytotoxic activities against MCF-7 cells (human breast cancer), with IC_{50} values of 19.4 and 12.5 μM, respectively [7].

Cholinesterase inhibitory activity

Compounds **95**, **104**, **151**, **156**, **342**, **352** and **353** were obtained from *A. bracteosa*, and their enzyme-inhibitory potential was evaluated. The diterpenoids **95**, **104**, **151** and **156** display inhibitory activity against cholinesterase (AChE and BChE) with IC_{50} values in the range of 14.0–35.2 μM for AChE and 10.0–19.0 μM for BChE, respectively. Compound **104** is the most active against cholinesterase while **156** is comparatively less active, indicating that the presence of a MeO group at C(15) increases the cholinesterase inhibitory activity [10].

Antioxidative activity

Bouderbala and co-workers studied the effect of *A. iva* aqueous extract on lipid peroxidation and antioxidant enzyme activity in hypercholesterolemic rats. The results showed that *A. iva* extract is more effective at improving RBC antioxidant capacity relative to that of tissues. In addition, *A. iva* aqueous extract can reduce oxidative stress, which may prevent lipid peroxidation in hypercholesterolemic models by increasing antioxidant enzyme activity [138].

Vasorelaxant activity

El-Hilaly and co-workers investigated vascular activity of *A. iva* aqueous extract in normotensive Wistar rats. The aqueous extract displayed NO-mediated and NO-independent vasorelaxing properties *in vitro*. The *A. iva* extract contains more than one active compound. One of these compounds is responsible for inhibition of noradrenaline

evoked contraction. Another compound was identified *in vitro* as a transient NO-dependent relaxation [8].

Conclusions

The plants of the genus *Ajuga* are widely distributed globally and many of these plants are used as traditional herbal medicines. The compounds isolated from this genus exert a broad spectrum of biological and pharmacological activities, however, our review indicates that phytochemical investigation has mainly focused on a few species. Further studies on the remaining species, their constituents and biological activities, should be carried out.

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