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Review

Chemical Constituents from *Croton* Species and Their Biological Activities

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Abstract: The genus *Croton* belongs to the Euphorbiaceae family, which comprises approximately 1300 species. Many *Croton* species have been used as folk medicines. This review focuses on the chemical constituents from *Croton* species and their relevant biological activities, covering the period from 2006 to 2018. A total of 399 new compounds, including 339 diterpenoids, were reported. Diterpenoids are characteristic components of the *Croton* species. These isolated compounds exhibited a broad spectrum of bioactivities, including cytotoxic, anti-inflammatory, antifungal, acetylcholinesterase inhibitory, and neurite outgrowth-promoting properties. The present review provides a significant clue for further research of the chemical constituents from the *Croton* species as potential medicines.

Keywords: *Croton* species; phytochemistry; biological activities; diterpenoids; cytotoxicity

1. Introduction

The genus *Croton* belongs to the Euphorbiaceae family, and contains approximately 1300 species of trees, shrubs, and herbs, which are widely distributed throughout tropical and subtropical regions of the world. Many *Croton* species have been used as folk medicines in Africa, south Asia, and south America, for the treatment of many diseases such as stomachache, abscesses, inflammation, and malaria [1–3]. The seeds of *C. tiglium*, which are well-known as "badou", had been utilized as a traditional Chinese medicine to treat gastrointestinal disorders, intestinal inflammation, and rheumatism. The roots of *C. crassifolius*, known as "jiguxiang" in China, are mainly used as a traditional medicine for the treatment of stomachache and sore throat [3]. The genus *Croton* is abundant in diverse diterpenoids, including clerodane, tigliane, kaurane, labdane, cembrane, and pimarane, with a wide range of biological activities, such as cytotoxic, anti-inflammatory, and anti-microbial [1–5]. Due to their great structural diversity and broad relevant bioactivities, *Croton* species have attracted increasing research attention. Several authors have provided reviews about the chemical constituents and biological activities of Croton species. A review came out in 2006 regarding clerodane diterpenes isolated from *Croton* species, their ^{13}C-NMR spectroscopic data, and biological activities [2]. In 2007, a comprehensive review on the traditional uses, chemistry, and pharmacology of *Croton* species was published [1]. In 2013, anticancer and antioxidant activities of extracts and pure compounds from several *Croton* species were reviewed [4]. Five review articles were published in recent years which focused on ethnopharmacological uses, phytochemistry, and pharmacology of a single *Croton* species [6–10]. In the last decade, there has been a dramatic progress in the chemical constituents and relevant biological activities of *Croton* species. However, so far, no comprehensive review has been published since 2007. In the present review, we summarize systematically the research advances on the new chemical constituents and their biological activities of *Croton* species reported in the literature,

as found on Web of Science, Google Scholar, PubMed, and SciFinder, from 2006 to March 2018, with the aim of providing a basis for further research of natural product drug discovery.

2. Chemical Constituents

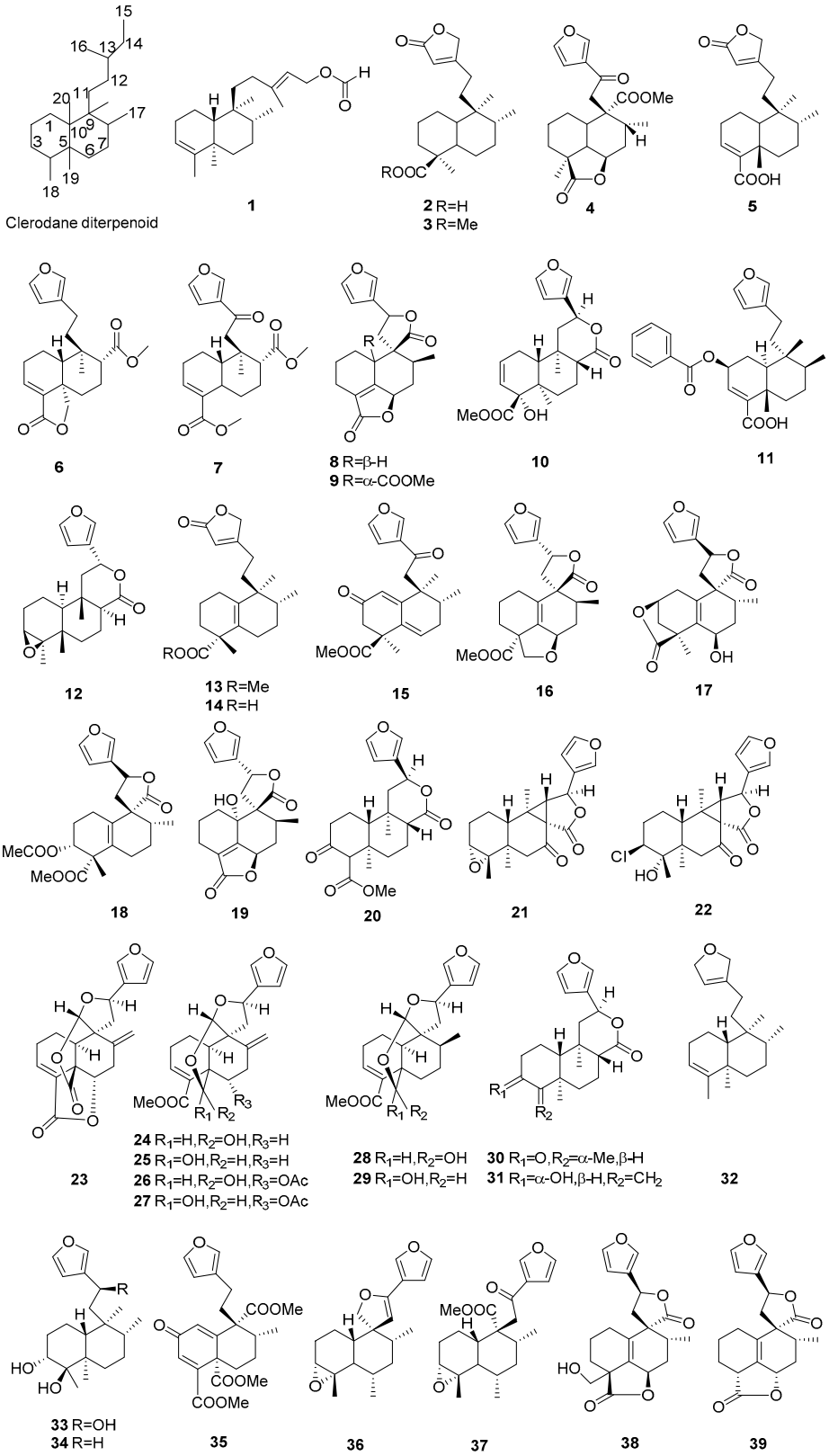
To date, 399 new compounds have been isolated and identified from *Croton* species, including 339 diterpenoids (**1**–**339**), seven sesquiterpenoids (**340**–**346**), one sesterterpenoid (**347**), one triterpenoid (**348**), 21 glycosides (**349**–**369**), eight alkaloids (**370**–**377**), three benzoate derivatives (**378**–**380**), three pyran-2-one derivatives (**381**–**383**), two cyclopeptide (**384**, **385**), two tropone derivatives (**386**, **387**), two limonoids (**388**, **389**), and ten miscellaneous compounds (**390**–**399**). Their structures, molecular formula, names, corresponding sources, and references are summarized in Figures 1–13 and Tables 1–27.

2.1. Diterpenoids

Phytochemical investigations on *Croton* species revealed the predominant secondary metabolites as diterpenoids, including clerodane, tigliane, kaurane, crotofolane, labdane, cembrane, abietane, casbane, halimane, pimarane, cleistanthane, grayanane, atisane, phytane, and laevinane diterpenoids. Three hundred & thirty-nine new diterpenoids (**1**–**339**) were reported from *Croton* species.

2.1.1. Clerodanes

Ninety-two new clerodane diterpenoids (**1**–**92**) were isolated from *Croton* species, including two clerodane diterpenoid with acyclic at C-9s, eight clerodane diterpenoids with butenolide at C-9, and 82 furan-clerodane diterpenoids [11]. Their structures, molecular formula, names, corresponding sources, and references are listed in Figure 1 and Table 1. Two new clerodane diterpenoids with acyclic side chain at C-9, *ent*-3,13*E*-clerodadiene-15-formate (**1**) and 3α,4α,15,16-tetrahydroxy-*ent*-*neo*-cleroda-13*E*-ene (**45**), were isolated from the roots of *C. sylvaticus* [12] and the roots of *C. limae* [13], respectively. Eight new clerodane diterpenoids with butenolide at C-9 (**2**, **3**, **5**, **13**, **14**, **75**, **91**, **92**) were obtained from three *Croton* species (*C. crassifolius*, *C. glabellus*, and *C. oligandrus*) [14–18]. Furan-clerodane diterpenoids are abundant in *Croton* species, and 82 new ones were isolated from different Croton species. For example, Centrafricine I (**4**) from *C. mayumbensis* was a new furan-clerodane diterpenoid with a 6, 18-γ-lactone ring [19]. Two novel rearranged *ent*-clerodane diterpenoids Laevinoids A, B (**21**, **22**) containing an unusual 3/5 bicyclic ring were obtained from the branches and leaves of *C. laevigatus*; **22** represents the first chlorinated example of the clerodane family [20]. Compounds (**23**–**27**) bearing a C-19/C-20 six-membered ring were identified from *C. laui* [21]. Phytochemical investigations on three *Croton* species (*C. oblongifolius*, *C. yanhuii*, and *C. hypoleucus*) afforded six new furan-clerodanoids (**12**, **36**, **37**, **84**–**86**) with a 3,4-epoxy moiety [22–24]. Crotoeurins A–C (**40**–**42**) were found from the twigs and leaves of *C. euryphyllus*. Among them, crotoeurin A (**40**) was a *nor*-clerodane diterpenoid dimer with a unique cyclobutane ring via a [2 + 2] cycloaddition [25]. Three new furan-clerodane diterpenoids, cracroson A–C (**46**–**48**) were obtained from *C. crassifolius*, while cracroson C (**48**) represents the first example of a clerodane diterpenoid alkaloid [26]. Twelve new *ent*-clerodanoids (**55**, **66**) were isolated from the roots of *C. megalocarpoides*. Among them, compounds (**58**–**66**) possessed 9, 12-γ-lactone ring [27]. Investigation on the roots of *C. crassifolius* afforded eight new clerodanoids, crassins A−H (**68**–**75**). Among them, crassins A–B (**68**, **69**) represents ring B rearranged clerodanoids, whereas crassins C (**70**) was ring A rearranged one [17]. One new *nor*-clerodane diterpenoid, norcrassifolin (**83**), with a 1,12-lactone six-membered ring, was isolated from *C. crassifolius* [28].



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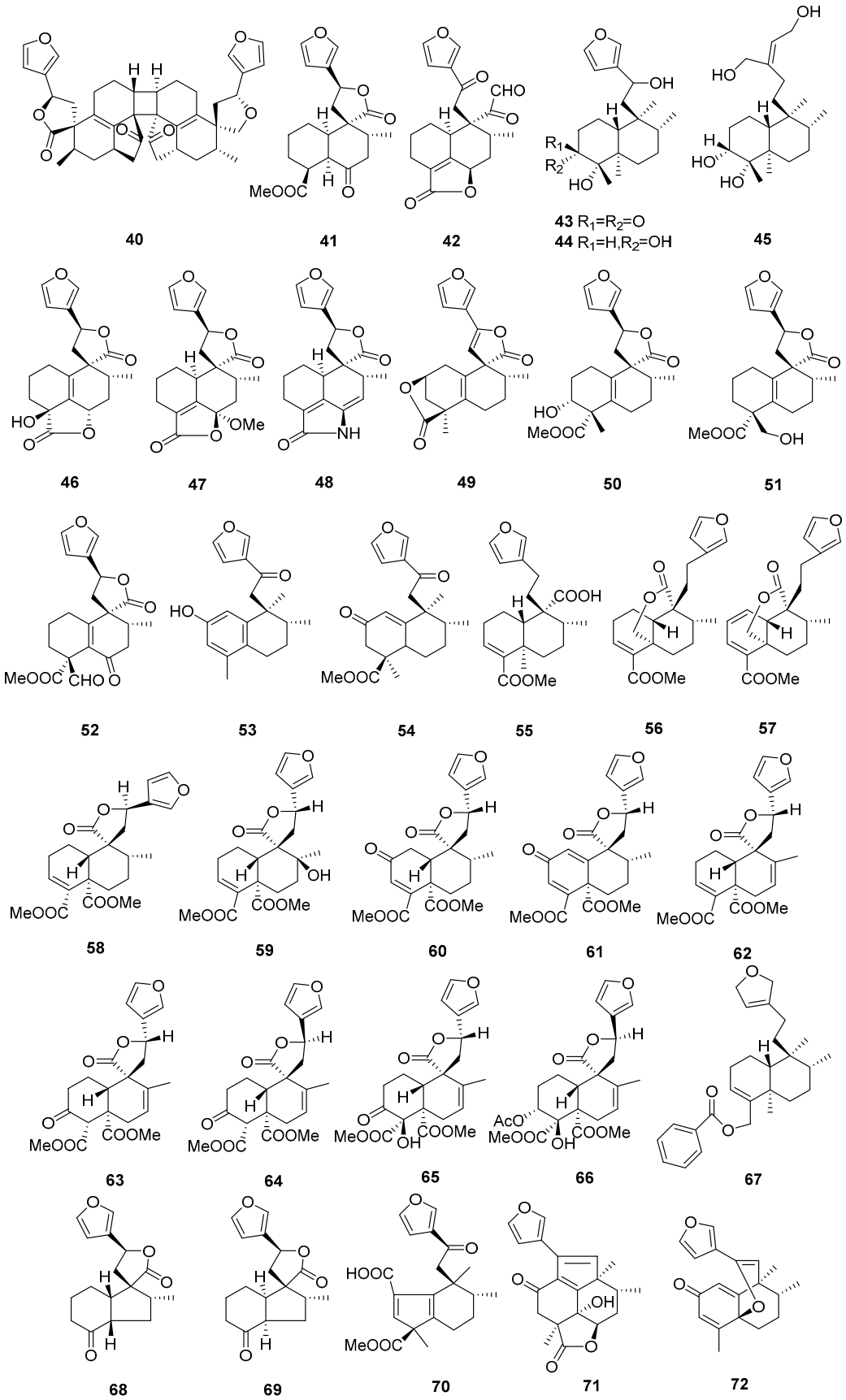

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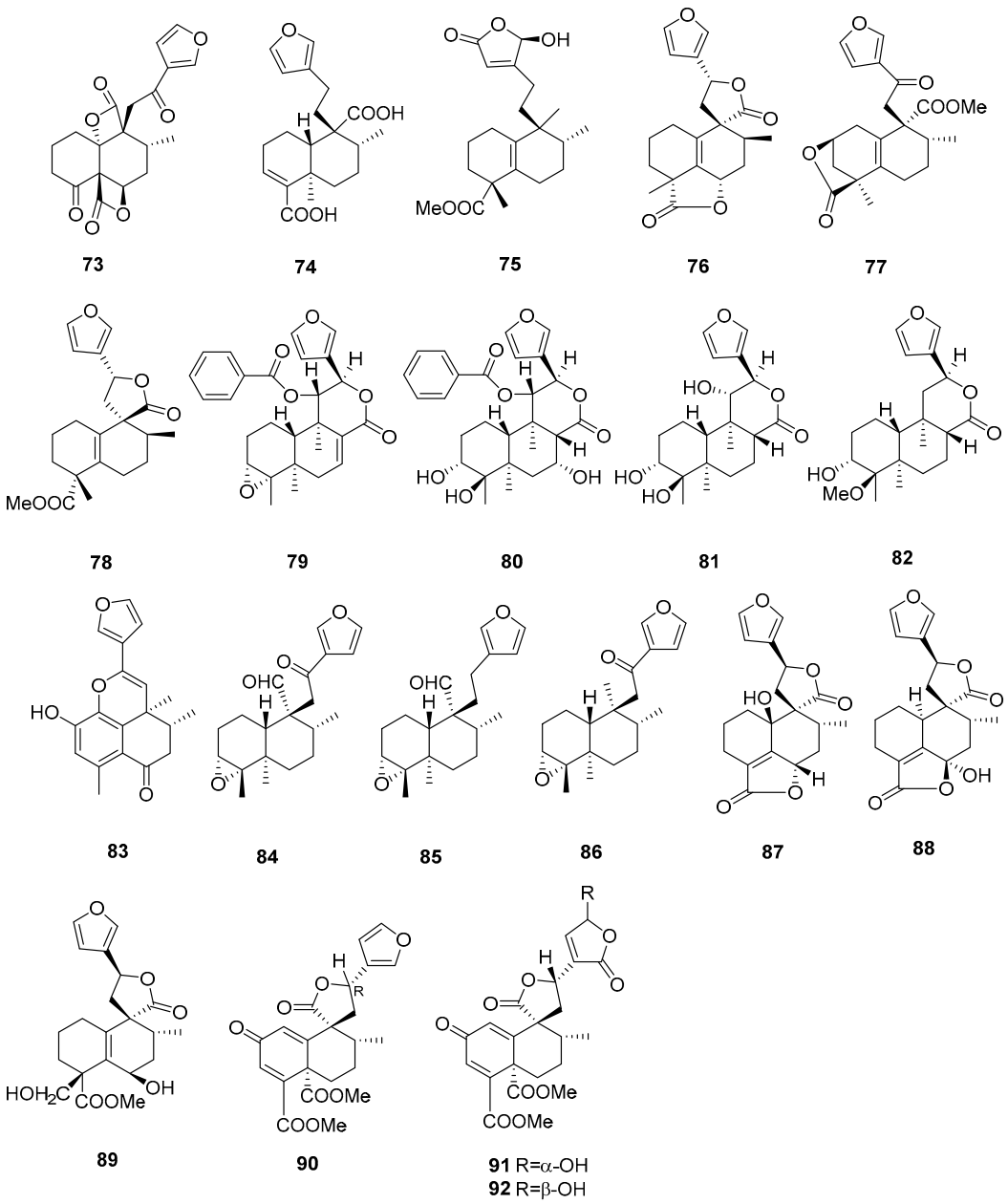

Figure 1. Clerodane type diterpenoids from the genus *Croton*.

Table 1. Clerodane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
1	*ent*-3,13*E*-clerodadiene-15-formate	$C_{21}H_{34}O_2$	*C. sylvaticus*	[12]
2	9-[2-(2(5*H*)-furanone-4-yl)ethyl]-4,8,9-trimethyl-1,2,3,4,5,6,7,8-octahydronaphthalene-4-carboxylic acid	$C_{20}H_{28}O_4$	*C. crassifolius*	[14]
3	9-[2-(2(5*H*)-furanone-4-yl)ethyl]-4,8,9-trimethyl-1,2,3,4,5,6,7,8-octahydronaphthalene-4-carboxylic ester	$C_{21}H_{30}O_4$	*C. crassifolius*	[14]
4	Centrafricine I	$C_{21}H_{24}O_6$	*C. mayumbensis*	[19]
5	Marrubiagenin	$C_{20}H_{28}O_4$	*C. glabellus*	[15]
6	Methyl 15,16-epoxy-3,13(16),14-*ent*-clerodatrien-18,19-olide-17-carboxylate	$C_{21}H_{26}O_5$	*C. oblongifolius*	[29]
7	Dimethyl 15,16-epoxy-12-oxo-3,13(16),14-*ent*-clerodatriene-17,18-dicarboxylate	$C_{22}H_{28}O_6$	*C. oblongifolius*	[29]
8	Isoteucvin	$C_{19}H_{20}O_5$	*C. jatrophoides*	[30]
9	Jatrophoidin	$C_{21}H_{22}O_7$	*C. jatrophoides*	[30]
10	8-Epicordatin	$C_{21}H_{26}O_6$	*C. palanostigma*	[31]
11	laevigatbenzoate	$C_{27}H_{31}O_5$	*C. laevigatus*	[13]
12	3,4,15,16-diepoxy-cleroda-13(16),14-diene-12,17-olide	$C_{20}H_{26}O_4$	*C. oblongifolius*	[22]
13	Crassifolin A	$C_{21}H_{30}O_4$	*C. crassifolius*	[16]
14	Crassifolin B	$C_{20}H_{29}O_4$	*C. crassifolius*	[16]
15	Crassifolin C	$C_{21}H_{24}O_5$	*C. crassifolius*	[16]
16	Crassifolin D	$C_{21}H_{24}O_6$	*C. crassifolius*	[16]
17	Crassifolin E	$C_{20}H_{23}O_6$	*C. crassifolius*	[16]
18	Crassifolin F	$C_{23}H_{29}O_7$	*C. crassifolius*	[16]
19	Crassifolin G	$C_{19}H_{20}O_6$	*C. crassifolius*	[16]
20	Methyl 3-oxo-12-epibarbascoate	$C_{21}H_{26}O_6$	*C. urucurana*	[32]
21	Laevinoids A	$C_{20}H_{22}O_5$	*C. laevigatus*	[20]
22	Laevinoids B	$C_{20}H_{23}O_5Cl$	*C. laevigatus*	[20]
23	Crotonolide A	$C_{20}H_{18}O_6$	*C. laui*	[21]
24	Crotonolide B	$C_{21}H_{24}O_6$	*C. laui*	[21]
25	Isocrotonolide B	$C_{21}H_{24}O_6$	*C. laui*	[21]
26	Crotonolide C	$C_{23}H_{26}O_8$	*C. laui*	[21]
27	Isocrotonolide C	$C_{23}H_{26}O_8$	*C. laui*	[21]
28	Crotonolide D	$C_{21}H_{26}O_6$	*C. laui*	[21]
29	Isocrotonolide D	$C_{21}H_{26}O_6$	*C. laui*	[21]
30	Crotonolide E	$C_{20}H_{26}O_4$	*C. laui*	[21]
31	Crotonolide F	$C_{20}H_{26}O_4$	*C. laui*	[21]
32	Crotonolide G	$C_{20}H_{32}O$	*C. laui*	[21]
33	Crotonolide H	$C_{20}H_{32}O_4$	*C. laui*	[21]
34	12-Deoxycrotonolide H	$C_{20}H_{32}O_3$	*C. laui*	[21]
35	Crotonoligaketone	$C_{23}H_{26}O_8$	*C. oligandrum*	[33]
36	Crotonpene A	$C_{20}H_{26}O_3$	*C. yanhuii*	[23]
37	Crotonpene B	$C_{21}H_{28}O_5$	*C. yanhuii*	[23]
38	Crassifolin I	$C_{20}H_{22}O_6$	*C. crassifolius*	[34]
39	Crassifolin H	$C_{19}H_{20}O_5$	*C. crassifolius*	[34]
40	Crotoeurin A	$C_{38}H_{36}O_1$	*C. euryphyllus*	[25]
41	Crotoeurin B	$C_{20}H_{24}O_6$	*C. euryphyllus*	[25]
42	Crotoeurin C	$C_{20}H_{22}O_6$	*C. euryphyllus*	[25]
43	3-Oxo-15,16-epoxy-4α,12-dihydroxy-*ent*-*neo*-clerodan-13(16),14-diene	$C_{20}H_{30}O_4$	*C. limae*	[35]

Table 1. *Cont.*

No.	Compound Name	Molecular Formula	Sources	Ref
44	15,16-Epoxy-3α,4α,12-trihydroxy-*ent*-*neo*-clerodan- 13(16),14-diene	$C_{20}H_{32}O_4$	*C. limae*	[35]
45	3α,4α,15,16-Tetrahydroxy-*ent*-*neo*-cleroda-13E-ene	$C_{20}H_{36}O_4$	*C. limae*	[35]
46	Cracroson A	$C_{19}H_{21}O_6$	*C. crassifolius*	[26]
47	Cracroson B	$C_{20}H_{22}O_6$	*C. crassifolius*	[26]
48	Cracroson C	$C_{19}H_{19}O_4N$	*C. crassifolius*	[26]
49	Crassifolin J	$C_{20}H_{20}O_5$	*C. crassifolius*	[36]
60	Crotocorylifuran-2-one	$C_{22}H_{24}O_8$	*C.megalocarpoides*	[27]
61	Megalocarpoidolide D	$C_{22}H_{22}O_8$	*C.megalocarpoides*	[27]
62	7,8-Dehydrocrotocorylifuran	$C_{22}H_{24}O_7$	*C.megalocarpoides*	[27]
63	Megalocarpoidolide E	$C_{22}H_{24}O_8$	*C.megalocarpoides*	[27]
64	Megalocarpoidolide F	$C_{22}H_{24}O_8$	*C.megalocarpoides*	[27]
65	Megalocarpoidolide G	$C_{22}H_{24}O_9$	*C.megalocarpoides*	[27]
66	Megalocarpoidolide H	$C_{24}H_{28}O_{10}$	*C.megalocarpoides*	[27]
67	Launine K	$C_{27}H_{36}O_3$	*C. laui*	[37]
68	Crassin A	$C_{17}H_{20}O_4$	*C. crassifolius*	[17]
69	Crassin B	$C_{17}H_{20}O_4$	*C. crassifolius*	[17]
70	Crassin C	$C_{21}H_{24}O_6$	*C. crassifolius*	[17]
71	Crassin D	$C_{20}H_{20}O_5$	*C. crassifolius*	[17]
72	Crassin E	$C_{19}H_{20}O_3$	*C. crassifolius*	[17]
73	Crassin F	$C_{19}H_{18}O_7$	*C. crassifolius*	[17]
74	Crassin G	$C_{20}H_{26}O_5$	*C. crassifolius*	[17]
75	Crassin H	$C_{21}H_{30}O_5$	*C. crassifolius*	[17]
76	Crassifolius A	$C_{20}H_{22}O_5$	*C. crassifolius*	[38]
77	Crassifolius B	$C_{21}H_{24}O_6$	*C. crassifolius*	[38]
78	Crassifolius C	$C_{21}H_{26}O_5$	*C. crassifolius*	[38]
79	Crolaevinoid C	$C_{27}H_{28}O_6$	*C. laevigatus*	[39]
80	Crolaevinoid D	$C_{27}H_{32}O_8$	*C. laevigatus*	[39]
81	Crolaevinoid E	$C_{20}H_{28}O_6$	*C. laevigatus*	[39]
82	Crolaevinoid F	$C_{21}H_{30}O_5$	*C. laevigatus*	[39]
83	Norcrassifolin	$C_{19}H_{18}O_4$	*C. crassifolius*	[28]
84	Hypolein A	$C_{20}H_{26}O_4$	*C. hypoleucus*	[24]
85	Hypolein B	$C_{20}H_{28}O_3$	*C. hypoleucus*	[24]
86	Hypolein C	$C_{20}H_{28}O_3$	*C. hypoleucus*	[24]
87	Cracroson E	$C_{19}H_{20}O_6$	*C. crassifolius*	[40]
88	Cracroson F	$C_{19}H_{20}O_6$	*C. crassifolius*	[40]
89	Cracroson G	$C_{21}H_{26}O_7$	*C. crassifolius*	[40]
90	12-Epi-megalocarpoidolide D	$C_{22}H_{22}O_8$	*C. oligandrus*	[18]
91	Crotonolins A	$C_{22}H_{22}O_{10}$	*C. oligandrus*	[18]
92	Crotonolins B	$C_{22}H_{22}O_{10}$	*C. oligandrus*	[18]

2.1.2. Tiglianes

Fifty-six new tigliane diterpenoids (**93**–**148**) were reported from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 2 and Table 2. Investigations on the aerial parts of *C. ciliatoglandulifer* produced four new tiglianoids (**95**–**98**). Among them, tiglianoids (**95**–**97**) possess a *N*,*N*-dimethyl moiety at 2′position [41]. Alienusolin (**107**) and compound (**111**) were obtained from the roots and the leaves of *C. alienus* and the leaves of *C. mauritianus*, respectively [42,43]. The twigs and leaves of *C. caudatus* produced three new tiglianoids, crotusins A–C (**128**–**130**) [44]. Tigliane diterpenoids were abundant in *C. tiglium*, other 47 new ones (**93**, **94**, **99**–**106**, **108**–**110**, **112**–**127**, **131**–**148**) were isolated from *C. tiglium* [45–52]. Among them, compound (**112**) was the first tiglianoid with the C20-aldehyde group [48].

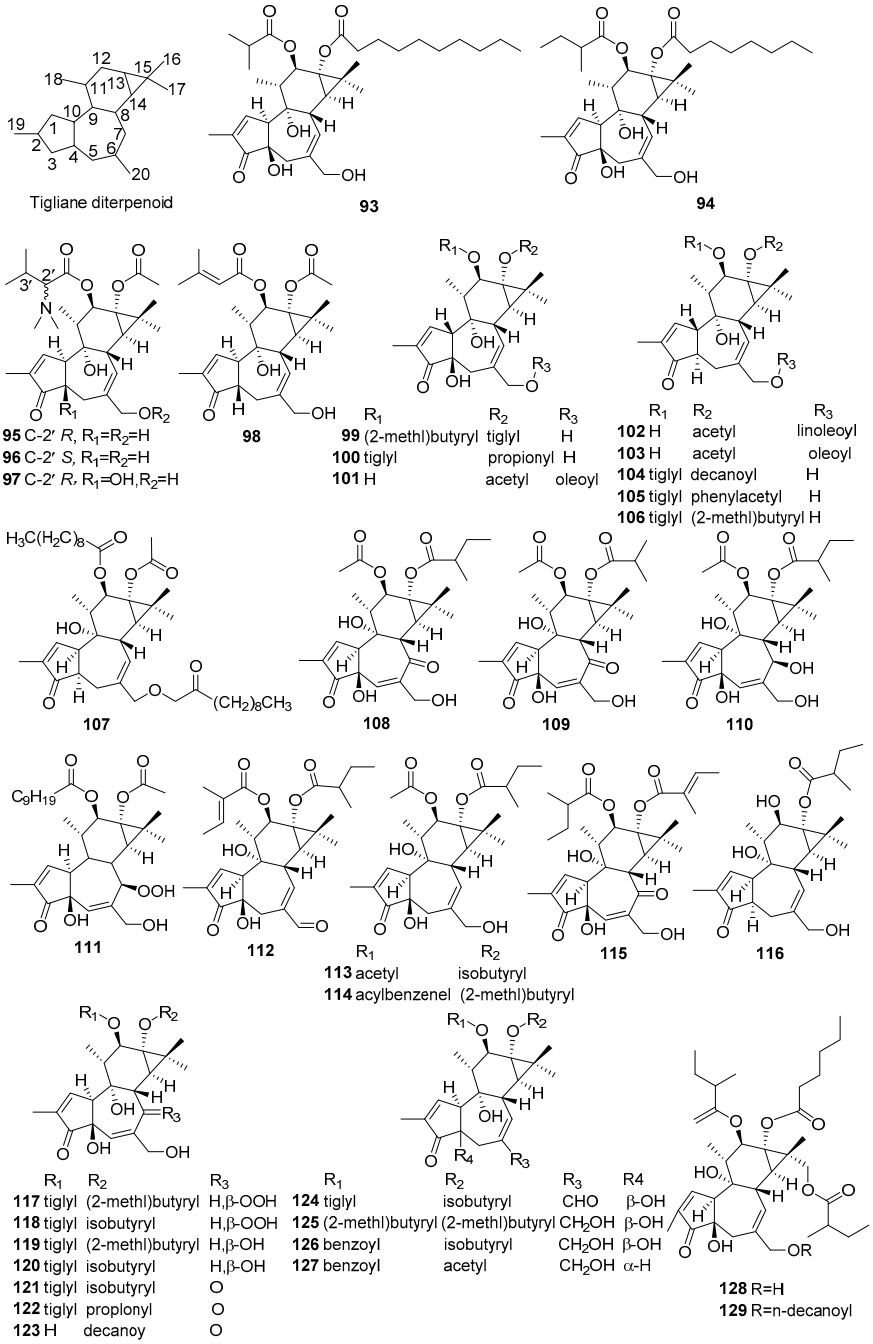

Figure 2. *Cont.*

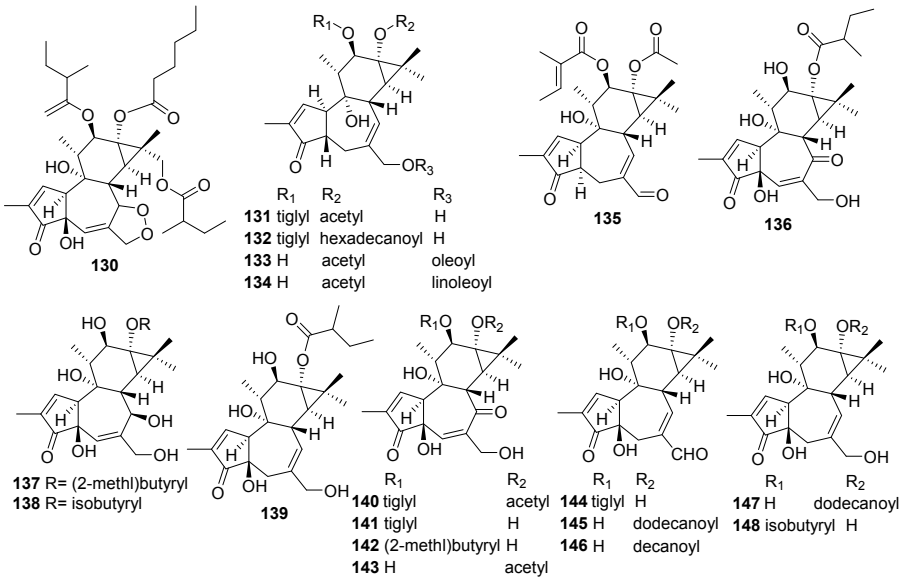

Figure 2. Tigliane type diterpenoids from the genus *Croton*.

Table 2. Tigliane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
93	12-*O*-isobutyrylphorbol-13-decanoate	$C_{34}H_{52}O_8$	*C. tiglium*	[45]
94	12-*O*-(2-methyl)butyrylphorbol-13-octanoate	$C_{33}H_{50}O_8$	*C. tiglium*	[45]
95	12-*O*-[(2R)-*N*,*N*-dimethyl-3-methylbutanoyl]-4-deoxyphorbol 13-acetate	$C_{29}H_{43}NO_7$	*C. ciliatoglandulifer*	[41]
96	12-*O*-[(2*S*)-*N*,*N*-dimethyl-3-methylbutanoyl]-4-deoxyphorbol 13-acetate	$C_{29}H_{43}NO_7$	*C. ciliatoglandulifer*	[41]
97	12-*O*-[(2*R*)-*N*,*N*-Dimethyl-3-methylbutanoyl]phorbol 13-acetate	$C_{29}H_{43}NO_8$	*C. ciliatoglandulifer*	[41]
98	12-*O*-[3-Methyl-2-butenoyl]-4-deoxyphorbol 13-acetate	$C_{27}H_{36}NO_7$	*C. ciliatoglandulifer*	[41]
99	12-*O*-(2-methyl)butyrylphorbol-13-tiglate	$C_{30}H_{42}O_8$	*C. tiglium*	[46]
100	12-*O*-tiglylphorbol-13-propionate	$C_{28}H_{38}O_8$	*C. tiglium*	[46]
101	13-*O*-acetylphorbol-20-oleate	$C_{40}H_{62}O_8$	*C. tiglium*	[46]
106	12-*O*-tiglyl-4-deoxy-4α-phorbol-13-(2-methyl)butyrate	$C_{30}H_{42}O_7$	*C. tiglium*	[46]
107	Alienusolin	$C_{42}H_{66}O_8$	*C. alienus*	[42]
108	12-*O*-acetyl-5,6-didehydro-7-oxophorbol-13-yl 2-methylbutanoate	$C_{27}H_{36}O_9$	*C. tiglium*	[47]
109	12-*O*-acetyl-5,6-didehydro-7-oxophorbol-13-yl2-methylpropanoate	$C_{26}H_{34}O_9$	*C. tiglium*	[47]
110	12-Oacetyl-5,6-didehydro-6,7-dihydro-7-hydroxyphorbol-13-yl 2-methylbutanoate	$C_{27}H_{38}O_9$	*C. tiglium*	[47]
111	12-*O*-decanoyl-7-hydroperoxy-phorbol-5-ene-13-acetate	$C_{32}H_{42}O_{10}$	*C. mauritianus*	[43]
112	20-deoxy-20-oxophorbol12-tiglate 13-(2-methyl)butyrate	$C_{30}H_{40}O_8$	*C. tiglium*	[48]
113	12-*O*-acetylphorbol-13-isobutyrate	$C_{26}H_{36}O_8$	*C. tiglium*	[48]
114	12-*O*-benzoylphorbol-13-(2-methyl)butyrate	$C_{32}H_{40}O_8$	*C. tiglium*	[48]
115	12-*O*-tiglyl-7-oxo-5-ene-phorbol-13-(2-methyl)butyrate	$C_{30}H_{40}O_9$	*C. tiglium*	[48]
116	13-*O*-(2-metyl)butyryl-4-deoxy-4a-phorbol	$C_{25}H_{36}O_6$	*C. tiglium*	[48]
117	Crotignoid A	$C_{30}H_{42}O_{10}$	*C. tiglium*	[49]
118	Crotignoid B	$C_{29}H_{40}O_{10}$	*C. tiglium*	[49]
119	Crotignoid C	$C_{30}H_{42}O_9$	*C. tiglium*	[49]
120	Crotignoid D	$C_{29}H_{40}O_9$	*C. tiglium*	[49]
121	Crotignoid E	$C_{29}H_{38}O_9$	*C. tiglium*	[49]
122	Crotignoid F	$C_{28}H_{36}O_9$	*C. tiglium*	[49]
123	Crotignoid G	$C_{30}H_{44}O_8$	*C. tiglium*	[49]
124	Crotignoid H	$C_{29}H_{38}O_8$	*C. tiglium*	[49]
125	Crotignoid I	$C_{30}H_{44}O_8$	*C. tiglium*	[49]
126	Crotignoid J	$C_{31}H_{38}O_8$	*C. tiglium*	[49]
127	Crotignoid K	$C_{29}H_{34}O_7$	*C. tiglium*	[49]
128	Crotusin A	$C_{36}H_{54}O_{10}$	*C. caudatus*	[44]
129	Crotusin B	$C_{46}H_{72}O_{11}$	*C. caudatus*	[44]
130	Crotusin C	$C_{36}H_{52}O_{11}$	*C. caudatus*	[44]
131	12-*O*-tiglylphorbol-4-deoxy- 4β-phorbol-13-acetate	$C_{27}H_{36}O_7$	*C. tiglium*	[50]
132	12-*O*-tiglylphorbol-4-deoxy-4β-phorbol-13-hexadecanoate	$C_{41}H_{64}O_7$	*C. tiglium*	[50]
133	13-*O*-acetylphorbol-4-deoxy-4β-phorbol-20-oleate	$C_{40}H_{62}O_7$	*C. tiglium*	[50]
134	13-*O*-acetylphorbol-4-deoxy-4β-phorbol-20-linoleate	$C_{40}H_{60}O_7$	*C. tiglium*	[50]
135	4-deoxy-20-oxophorbol 12-tiglyl 13-acetate	$C_{27}H_{34}O_7$	*C. tiglium*	[51]
136	7-oxo-5-ene-phorbol-13-(2-methylbutyrate)	$C_{25}H_{34}O_8$	*C. tiglium*	[51]
137	7-hydroxyl-phorbol-5-ene-13-(2-methyl)butyrate	$C_{25}H_{36}O_8$	*C. tiglium*	[51]

2.1.3. Kauranes

Fourty new kaurane diterpenoids (**149**–**188**) were found from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are listed in Figure 3 and Table 3. Five new 3,4-*seco ent*-kauranes (**149**–**150**, **160**–**161**, **168**) were isolated from *C. caracasana* [53],

C. megistocarpus [54], and *C. oblongifolius* [55], respectively. Investigations on *C. kongensis* afforded eight new 8,9-*seco*-*ent*-kaurane diterpenes (**151**–**154**, **156**, **178**–**180**) [56–59]. Compound **181**, one new kaurane bearing a monoterpene unit at C-16, was found from *C. limae* [35]. From the stems of *C. micans*, five new 3,4-*seco*-*ent*-kaurene dimers (**182**–**186**) were isolated [60], while other two dimeric *ent*-kaurane diterpenoids (**187**–**188**) were elucidated from *C. tonkinensis* [61].

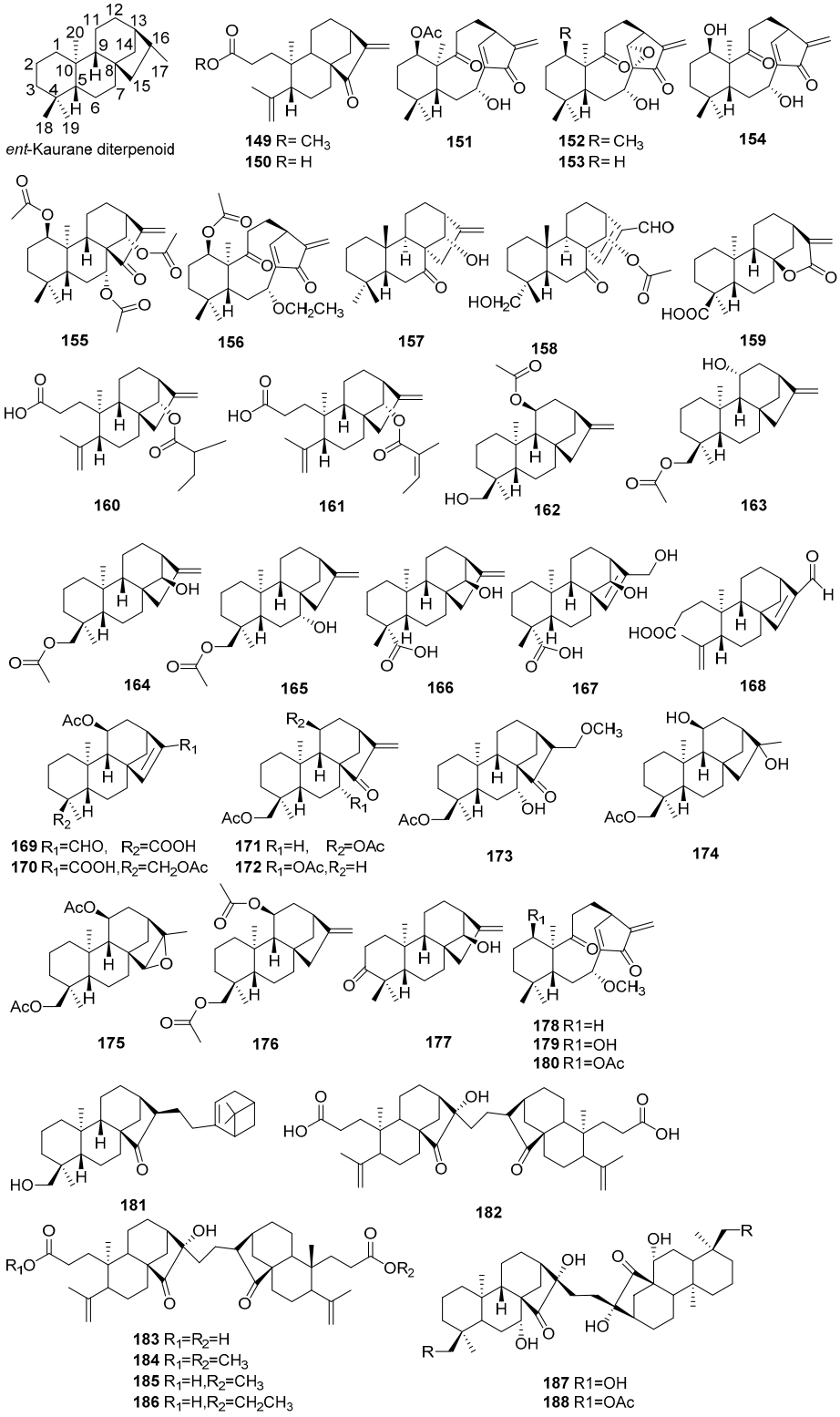

Figure 3. Kaurane type diterpenoids from the genus *Croton* 1.

Table 3. Kaurane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
149	Caracasine	$C_{21}H_{30}O_3$	*C. caracasana*	[53]
150	Caracasine acid	$C_{20}H_{28}O_3$	*C. caracasana*	[53]
151	Kongensin A	$C_{22}H_{30}O_5$	*C. kongensis*	[56]
152	Kongensin B	$C_{22}H_{30}O_6$	*C. kongensis*	[56]
153	Kongensin C	$C_{20}H_{28}O_5$	*C. kongensis*	[56]
154	Kongensin D	$C_{20}H_{28}O_4$	*C. kongensis*	[57]
155	Kongensin E	$C_{26}H_{36}O_7$	*C. kongensis*	[57]
156	Kongensin F	$C_{24}H_{34}O_5$	*C. kongensis*	[58]
157	Crotonkinin A	$C_{20}H_{30}O_2$	*C. tonkinensis*	[62]
158	Crotonkinin B	$C_{22}H_{32}O_4$	*C. tonkinensis*	[62]
159	14-*epi*-hyalic acid	$C_{20}H_{28}O_4$	*C. argyrophylloides*	[63]
160	14-[(2-methylbutanoyl)oxy]-3,4-seco-*ent*-kaura-4(19),16-dien-3-oic acid	$C_{25}H_{39}O_4$	*C. megistocarpus*	[54]
161	14-{[(2*Z*)-2-methylbut-2-enoyl]oxy}-3,4-seco-*ent*-kaura-4(19),16-dien-3-oic acid	$C_{25}H_{37}O_4$	*C. megistocarpus*	[54]
162	*ent*-11β-acetoxykaur-16-en-18-ol	$C_{22}H_{34}O_3$	*C. tonkinensis*	[64]
163	*ent*-11α-hydroxy-18-acetoxykaur-16-ene	$C_{22}H_{34}O_3$	*C. tonkinensis*	[64]
164	*ent*-14β-hydroxy-18-acetoxykaur-16-ene	$C_{22}H_{34}O_3$	*C. tonkinensis*	[64]
165	*ent*-7α-hydroxy-18-acetoxykaur-16-ene	$C_{22}H_{34}O_3$	*C. tonkinensis*	[64]
166	*ent*-14S*-hydroxykaur-16-en-19-oic acid	$C_{20}H_{30}O_3$	*C. pseudopulchellus*	[65]
167	*ent*-14S*,17-dihydroxykaur-15-en-19-oic acid	$C_{20}H_{30}O_4$	*C. pseudopulchellus*	[65]
168	*ent*-3,4-*seco*-17-oxo-kaur-4(19),15(16)-dien-3-oic acid	$C_{20}H_{28}O_3$	*C. oblongifolius*	[55]
169	Crotonkinin C	$C_{22}H_{30}O_5$	*C. tonkinensis*	[66]
170	Crotonkinin D	$C_{24}H_{34}O_6$	*C. tonkinensis*	[66]
171	Crotonkinin E	$C_{24}H_{34}O_5$	*C. tonkinensis*	[66]
172	Crotonkinin F	$C_{24}H_{34}O_5$	*C. tonkinensis*	[66]
173	Crotonkinin G	$C_{23}H_{36}O_5$	*C. tonkinensis*	[66]
174	Crotonkinin H	$C_{22}H_{36}O_4$	*C. tonkinensis*	[66]
175	Crotonkinin I	$C_{24}H_{36}O_5$	*C. tonkinensis*	[66]
176	Crotonkinin J	$C_{23}H_{34}O_5$	*C. tonkinensis*	[66]
177	14β-hydroxy-3-oxo-ent-kaur-16-ene	$C_{20}H_{30}O_2$	*C. kongensis*	[67]
178	Kongeniod A	$C_{21}H_{30}O_3$	*C. kongensis*	[59]
179	Kongeniod B	$C_{21}H_{30}O_4$	*C. kongensis*	[59]
180	Kongeniod C	$C_{23}H_{32}O_5$	*C. kongensis*	[59]
181	15-oxo-17(10′-α-pinenyl)-kauran-18-oic acid	$C_{30}H_{44}O_3$	*C. limae*	[35]
182	Micansinoic acid	$C_{40}H_{58}O_7$	*C. micans*	[60]
183	Isomicansinoic acid	$C_{40}H_{58}O_7$	*C. micans*	[60]
184	Dimethylester of micansinoic	$C_{42}H_{62}O_7$	*C. micans*	[60]
185	Methyl-micansinoic acid	$C_{41}H_{60}O_7$	*C. micans*	[60]
186	Ethyl-micansinoic acid	$C_{42}H_{62}O_7$	*C. micans*	[60]
187	Crotonkinensin C	$C_{40}H_{62}O_8$	*C. tonkinensis*	[61]
188	Crotonkinensin D	$C_{44}H_{66}O_{10}$	*C. tonkinensis*	[61]

2.1.4. Crotofolanes

Thirty-nine new crotofolane diterpenoids (**189**–**227**) were obtained from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are summarized in Figure 4 and Table 4. Twenty-four new crotofolane diterpenoids (**189**–**198**, **212**–**222**, **225**–**227**) were isolated from *C. caracasanus* [68–71]. Among them, three new crotofolane diterpenoid alkaloids, cascarinoids A–C (**225**–**227**), were firstly found. Investigations on *C. argyrophyllus* gave four new crotofolanes (**199**–**202**) [72]. Crotocarasin A–D (**203**–**206**) were isolated from the stems of *C. caracasanus* [73]. Five new 1, 14-*seco*-crotofolanes from *C. insularis* were obtained [74], while *C. dichogamus* yielded crotodichogamoin A–B (**223**–**224**) [75].

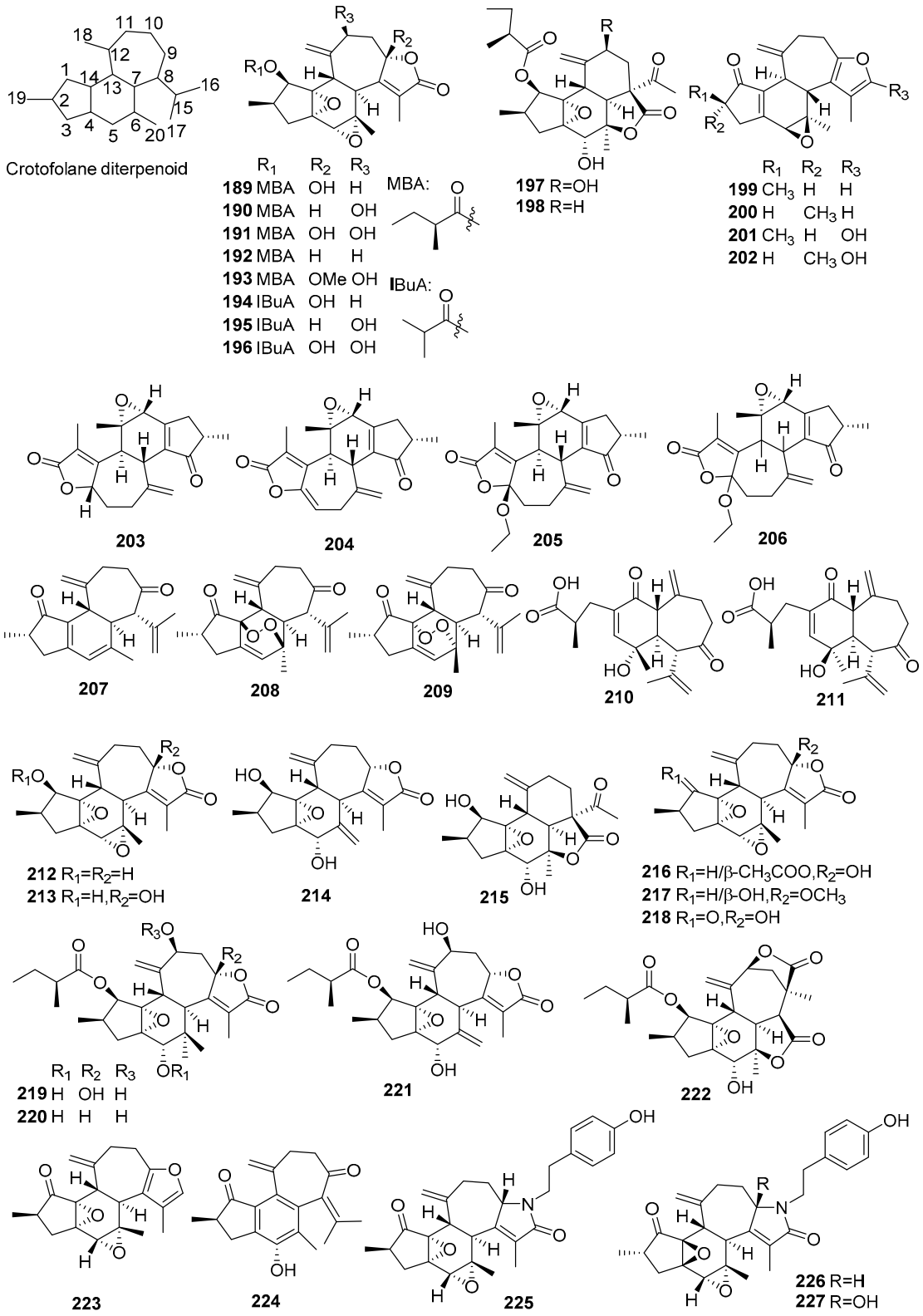

Figure 4. Crotofolane type diterpenoids from the genus *Croton*.

Table 4. Crotofolane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
189	Crotocascarin A	$C_{25}H_{32}O_7$	*C. cascarilloides*	[68]
190	Crotocascarin B	$C_{25}H_{32}O_7$	*C. cascarilloides*	[68]
191	Crotocascarin C	$C_{25}H_{32}O_8$	*C. cascarilloides*	[68]
192	Crotocascarin D	$C_{25}H_{32}O_6$	*C. cascarilloides*	[68]
193	Crotocascarin E	$C_{26}H_{34}O_8$	*C. cascarilloides*	[68]
194	Crotocascarin F	$C_{24}H_{30}O_7$	*C. cascarilloides*	[68]
195	Crotocascarin G	$C_{24}H_{30}O_7$	*C. cascarilloides*	[68]
196	Crotocascarin H	$C_{24}H_{30}O_8$	*C. cascarilloides*	[68]
197	Crotocascarin α	$C_{24}H_{32}O_8$	*C. cascarilloides*	[68]
198	Crotocascarin β	$C_{24}H_{32}O_7$	*C. cascarilloides*	[68]
199	(5β,6β)-5,6: 13,16-diepoxycrotofola-4(9),10(18),13,15-tetraen-1-one	$C_{20}H_{22}O_3$	*C. argyrophyllus*	[72]
200	(5β,6β)-5,6: 13,16-diepoxy-2-epicrotofola-4(9),10(18),13,15-tetraen-1-one	$C_{20}H_{22}O_3$	*C. argyrophyllus*	[72]
201	(5β,6β)-5,6: 13,16-diepoxy-16-hydroxycrotofola-4(9),10(18),13,15-tetraen-1-one	$C_{20}H_{22}O_4$	*C. argyrophyllus*	[72]
202	(5β,6β)-5,6: 13,16-diepoxy-16-hydroxy-2-epi-crotofola-4(9),10(18),13,15-tetraen-1-one	$C_{20}H_{22}O_4$	*C. argyrophyllus*	[72]
203	Crotocarasin A	$C_{20}H_{22}O_4$	*C. caracasanus*	[73]
204	Crotocarasin B	$C_{20}H_{22}O_4$	*C. caracasanus*	[73]
205	Crotocarasin C	$C_{22}H_{26}O_5$	*C. caracasanus*	[73]
206	Crotocarasin D	$C_{22}H_{26}O_5$	*C. caracasanus*	[73]
207	EBC-162	$C_{20}H_{24}O_2$	*C. insularis*	[74]
208	EBC-233	$C_{20}H_{24}O_4$	*C. insularis*	[74]
209	EBC-300	$C_{20}H_{24}O_4$	*C. insularis*	[74]
210	EBC-240	$C_{20}H_{26}O_5$	*C. insularis*	[74]
211	EBC-241	$C_{20}H_{26}O_5$	*C. insularis*	[74]
212	Crotocascarin I	$C_{20}H_{24}O_5$	*C. cascarilloides*	[69]
213	Crotocascarin J	$C_{20}H_{24}O_6$	*C. cascarilloides*	[69]
214	Crotocascarin K	$C_{20}H_{24}O_5$	*C. cascarilloides*	[69]
215	Crotocascarin γ	$C_{19}H_{24}O_6$	*C. cascarilloides*	[69]
216	Crotocascarin L	$C_{22}H_{26}O_7$	*C. cascarilloides*	[70]
217	Crotocascarin M	$C_{21}H_{26}O_6$	*C. cascarilloides*	[70]
218	Crotocascarin N	$C_{20}H_{22}O_6$	*C. cascarilloides*	[70]
219	Crotocascarin O	$C_{25}H_{34}O_9$	*C. cascarilloides*	[70]
220	Crotocascarin P	$C_{25}H_{34}O_8$	*C. cascarilloides*	[70]
221	Crotocascarin Q	$C_{25}H_{32}O_7$	*C. cascarilloides*	[70]
222	Neocrotocascarin	$C_{25}H_{32}O_8$	*C. cascarilloides*	[70]
223	Crotodichogamoin A	$C_{20}H_{22}O_4$	*C. dichogamus*	[75]
224	Crotodichogamoin B	$C_{20}H_{22}O_2$	*C. dichogamus*	[75]
225	Cascarinoid A	$C_{28}H_{31}NO_5$	*C. cascarilloides*	[71]
226	Cascarinoid B	$C_{28}H_{31}NO_5$	*C. cascarilloides*	[71]
227	Cascarinoid C	$C_{28}H_{31}NO_6$	*C. cascarilloides*	[71]

2.1.5. Labdanes

Thirty-six new labdane diterpenoids (**228**–**263**) were isolated from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 5 and Table 5 12 new labdanes (**228**, **249**–**259**) were isolated from *C. laui* [21,76,77]. From the leaves of *C. stipuliformis*, three 3,4-*seco*-*ent*-labdanes (**229**–**231**) and one *ent*-labdane (**232**) were obtained [78]. Investigation of *C. laevigatus* led to the isolation of 16 new labdanes (**233**–**248**). Among them, crotonlaevins A–B (**233**, **234**), represents rare labdanes with a dodecahydronaphtho [1,2-c] furan moiety [79]. Three new labdane diterpenoids (**260**–**262**) were found from *C. jacobinensis* [77] and *C. decalvatus* [80], respectively. Bicrotonol A (**263**), one dimeric labdane-type diterpenoid, was obtained from the roots of *C. crassifolius* [81].

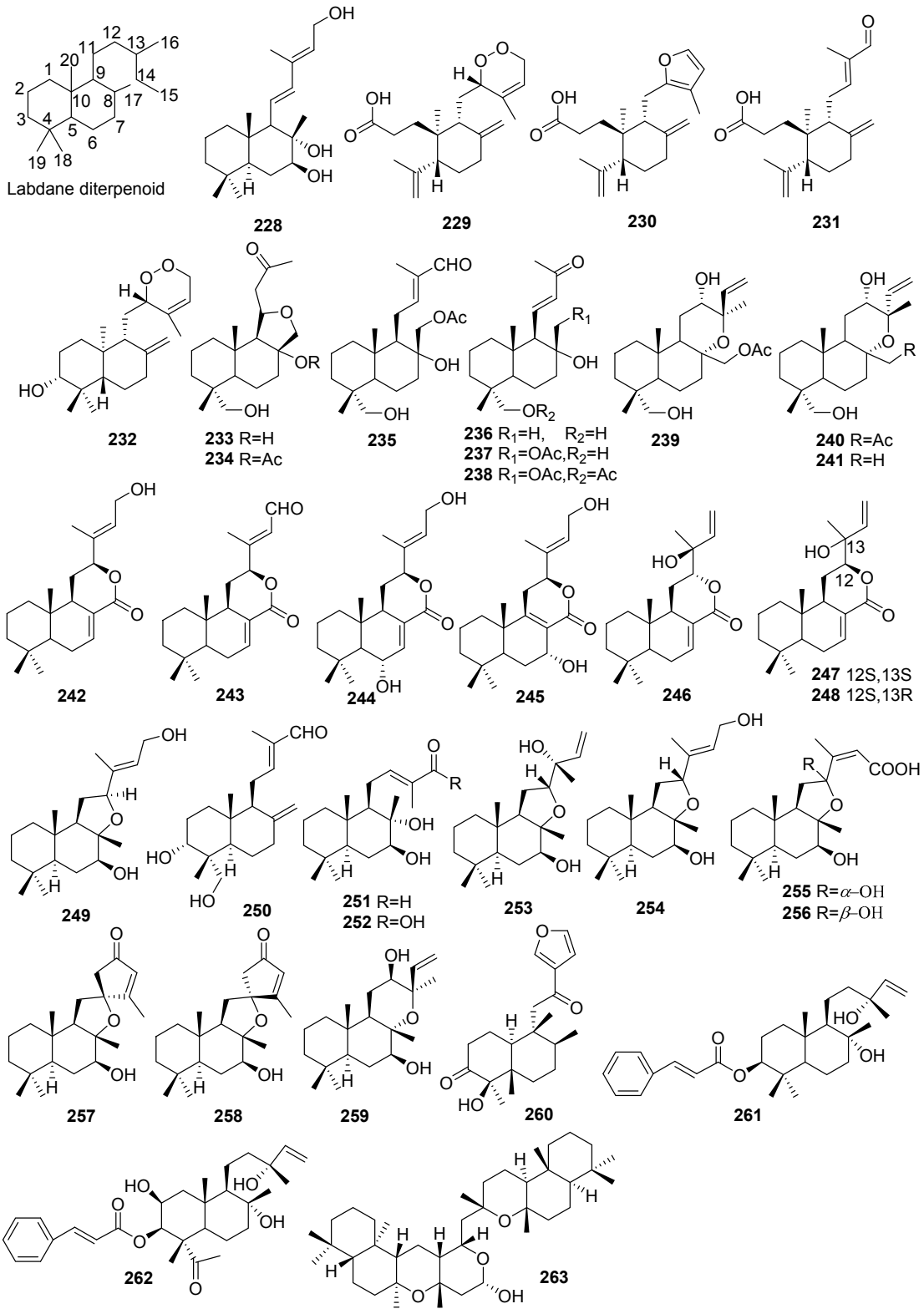

Figure 5. Labdane type diterpenoids from the genus *Croton*.

Table 5. Labdane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
228	Labdinine N	$C_{20}H_{34}O_3$	*C. laui*	[76]
229	*ent*-12,15-dioxo-3,4-*seco*-4,8,13-labdatrien-3-oic acid	$C_{20}H_{28}O_4$	*C. stipuliformis*	[78]
230	*ent*-12,15-epoxy-3,4-*seco*-4,8,12,14-labdatetraen-3-oic acid	$C_{20}H_{28}O_3$	*C. stipuliformis*	[78]
231	*ent*-15-nor-14-oxo-3,4-seco-4,8,12(*E*)-labdatrien-3-oic acid	$C_{19}H_{28}O_3$	*C. stipuliformis*	[78]
232	*ent*-12,15-dioxo-8,13-labdadien-3*a*-ol	$C_{20}H_{28}O_3$	*C. stipuliformis*	[78]
233	Crotonlaevin A	$C_{18}H_{30}O_4$	*C. laevigatus*	[79]
234	Crotonlaevin B	$C_{20}H_{32}O_5$	*C. laevigatus*	[79]
235	Crotonlaevin C	$C_{21}H_{34}O_5$	*C. laevigatus*	[79]
236	Crotonlaevin D	$C_{18}H_{30}O_3$	*C. laevigatus*	[79]
237	Crotonlaevin E	$C_{20}H_{32}O_5$	*C. laevigatus*	[79]
238	Crotonlaevin F	$C_{22}H_{34}O_6$	*C. laevigatus*	[79]
239	Crotonlaevin G	$C_{22}H_{36}O_5$	*C. laevigatus*	[79]
240	Crotonlaevin H	$C_{22}H_{36}O_5$	*C. laevigatus*	[79]
241	Crotonlaevin I	$C_{20}H_{34}O_4$	*C. laevigatus*	[79]
242	Crotonlaevin J	$C_{20}H_{30}O_3$	*C. laevigatus*	[79]
243	Crotonlaevin K	$C_{20}H_{28}O_3$	*C. laevigatus*	[79]
244	Crotonlaevin L	$C_{20}H_{30}O_4$	*C. laevigatus*	[79]
245	Crotonlaevin M	$C_{20}H_{30}O_4$	*C. laevigatus*	[79]
246	Crotonlaevin N	$C_{20}H_{30}O_3$	*C. laevigatus*	[79]
247	Crotonlaevin O	$C_{20}H_{30}O_3$	*C. laevigatus*	[79]
248	Crotonlaevin P	$C_{20}H_{30}O_3$	*C. laevigatus*	[79]
249	Crotonolide I	$C_{20}H_{34}O_3$	*C. laui*	[21]
250	Crotonolide J	$C_{19}H_{30}O_3$	*C. laui*	[21]
251	Launine A	$C_{19}H_{32}O_3$	*C. laui*	[82]
252	Launine B	$C_{19}H_{32}O_4$	*C. laui*	[82]
253	Launine C	$C_{20}H_{34}O_3$	*C. laui*	[82]
254	Launine D	$C_{20}H_{34}O_3$	*C. laui*	[82]
255	Launine E	$C_{20}H_{32}O_5$	*C. laui*	[82]
256	Launine F	$C_{20}H_{32}O_5$	*C. laui*	[82]
257	Launine G	$C_{20}H_{30}O_4$	*C. laui*	[82]
258	Launine H	$C_{20}H_{30}O_4$	*C. laui*	[82]
259	Launine I	$C_{20}H_{34}O_3$	*C. laui*	[82]
260	15,16-epoxy-4-hydroxy-labda-13(16),14-dien-3,12-dione	$C_{20}H_{28}O_4$	*C. jacobinensis*	[77]
261	Crotondecalvatin A	$C_{29}H_{42}O_4$	*C. decalvatus*	[80]
262	Crotondecalvatin B	$C_{30}H_{42}O_6$	*C. decalvatus*	[80]
263	Bicrotonol A	$C_{40}H_{68}O_4$	*C. crassifolius*	[81]

2.1.6. Cembranes

A total of 28 new cembrane diterpenoids (**264**–**291**) were obtained from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are listed in Figure 6 and Table 6. launine O-P (**264**, **265**), two new cembranes, were reported from the aerial parts of *C. laui* [76]. Investigations on the stem bark of *C. oblongifolius* afforded four new furanocembranoids (**266**–**269**) [83]. laevigatlactones A–F (**270**–**275**), six new cembranoids possessing a rare six-membered lactone moiety attached to C-1 and C-20, were firstly isolated from *C. laevigatus* [84]. 14 new cembranoids (**276**–**289**) were found from *C. gratissimus* [85,86]. Among them, compound **276** was first example of a 2,12-cyclocembranolide. The leaves of *C. longissimus* produced two new cembranes (**290**, **291**) [87].

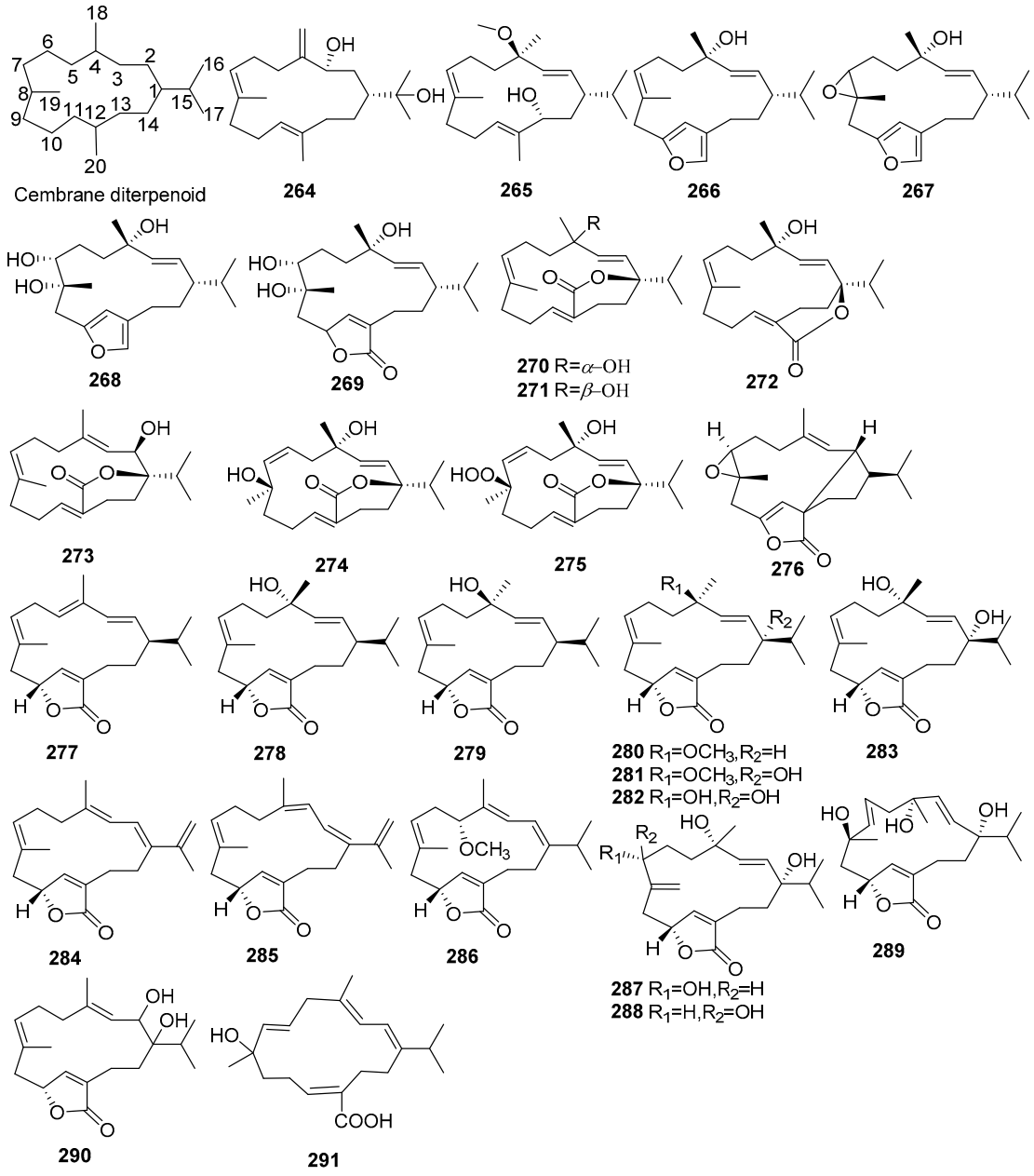

Figure 6. Cembrane type diterpenoids from the genus *Croton*.

Table 6. Cembrane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
264	Launine O	$C_{20}H_{34}O_2$	*C. laui*	[76]
265	Launine P	$C_{21}H_{36}O_2$	*C. laui*	[76]
266	Furanocembranoid 1	$C_{20}H_{30}O_2$	*C. oblongifolius*	[83]
267	Furanocembranoid 2	$C_{20}H_{30}O_3$	*C. oblongifolius*	[83]
268	Furanocembranoid 3	$C_{20}H_{32}O_4$	*C. oblongifolius*	[83]
269	Furanocembranoid 4	$C_{20}H_{32}O_5$	*C. oblongifolius*	[83]
270	Laevigatlactone A	$C_{20}H_{30}O_3$	*C. laeVigatus*	[84]
271	Laevigatlactone C	$C_{20}H_{30}O_3$	*C. laeVigatus*	[84]
272	Laevigatlactone B	$C_{20}H_{30}O_3$	*C. laeVigatus*	[84]
273	Laevigatlactone D	$C_{20}H_{30}O_3$	*C. laeVigatus*	[84]
274	Laevigatlactone E	$C_{20}H_{30}O_4$	*C. laeVigatus*	[84]
275	Laevigatlactone F	$C_{20}H_{30}O_5$	*C. laeVigatus*	[84]
276	(+)-[1R*,2S*,7S*,8S*,12R*]-7,8-Epoxy-2,12-cyclocembra-3*E*,10*Z*dien-20,10-olide	$C_{20}H_{28}O_3$	*C. gratissimus*	[85]
277	(+)-[1R*,10R*]-Cembra-2*E*,4*E*,7*E*,11*Z*-tetraen-20,10-olide	$C_{20}H_{28}O_2$	*C. gratissimus*	[85]
278	(+)-[1R*,4S*,10R*]-4-Hydroxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_3$	*C. gratissimus*	[85]
279	(−)-[1R*,4R*,10R*]-4-Hydroxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_3$	*C. gratissimus*	[85]
280	(−)-(1R*,4R*,10R*)-4-Methoxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide	$C_{21}H_{32}O_3$	*C. gratissimus*	[86]
281	(−)-(1S*,4R*,10R*)-1-Hydroxy-4-methoxycembra-2*E*,7*E*,11*Z*trien-20,10-olide	$C_{21}H_{32}O_4$	*C. gratissimus*	[86]
282	(−)-(1S*,4S*,10R*)-1,4-Dihydroxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_4$	*C. gratissimus*	[86]
283	(−)-(1S*,4S*,10R*)-1,4-Dihydroxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_4$	*C. gratissimus*	[86]
284	(+)-(10R*)-Cembra-1*E*,3*E*,7*E*,11*Z*,16-pentaen-20,10-olide	$C_{20}H_{26}O$	*C. gratissimus*	[86]
285	(+)-(10R*)-Cembra-1*Z*,3*Z*,7*E*,11*Z*,15-pentaen-20,10-olide	$C_{20}H_{26}O$	*C. gratissimus*	[86]
286	(+)-(5R*,10R*)-5-Methoxycembra-1*E*,3*E*,7*E*,11*Z*,15-pentaen-20,10-olide	$C_{21}H_{30}O_3$	*C. gratissimus*	[86]
287	(+)-(1S*,4S*,7R*,10R*)-1,4,7-Trihydroxycembra-2*E*,8(19),11*Z*-trien-20,10-olide	$C_{20}H_{30}O_5$	*C. gratissimus*	[86]
288	(−)-(1S*,4S*,7S*,10R*)-1,4,7-Trihydroxycembra-2*E*,8(19),11*Z*-trien-20,10-olide	$C_{20}H_{30}O_3$	*C. gratissimus*	[86]
289	(+)-(1S*,4R*,8S*,10R*)-1,4,8-Trihydroxycembra-2*E*,6*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_5$	*C. gratissimus*	[86]
290	Cembranoid 1	$C_{20}H_{30}O_4$	*C. longissimus*	[87]
291	Cembranoid 2	$C_{20}H_{30}O_3$	*C. longissimus*	[87]
281	(−)-(1S*,4R*,10R*)-1-Hydroxy-4-methoxycembra-2*E*,7*E*,11*Z*trien-20,10-olide	$C_{21}H_{32}O_4$	*C. gratissimus*	[86]
282	(−)-(1S*,4S*,10R*)-1,4-Dihydroxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_4$	*C. gratissimus*	[86]
283	(−)-(1S*,4S*,10R*)-1,4-Dihydroxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_4$	*C. gratissimus*	[86]
284	(+)-(10R*)-Cembra-1*E*,3*E*,7*E*,11*Z*,16-pentaen-20,10-olide	$C_{20}H_{26}O$	*C. gratissimus*	[86]
285	(+)-(10R*)-Cembra-1*Z*,3*Z*,7*E*,11*Z*,15-pentaen-20,10-olide	$C_{20}H_{26}O$	*C. gratissimus*	[86]
286	(+)-(5R*,10R*)-5-Methoxycembra-1*E*,3*E*,7*E*,11*Z*,15-pentaen-20,10-olide	$C_{21}H_{30}O_3$	*C. gratissimus*	[86]
287	(+)-(1S*,4S*,7R*,10R*)-1,4,7-Trihydroxycembra-2*E*,8(19),11*Z*-trien-20,10-olide	$C_{20}H_{30}O_5$	*C. gratissimus*	[86]
288	(−)-(1S*,4S*,7S*,10R*)-1,4,7-Trihydroxycembra-2*E*,8(19),11*Z*-trien-20,10-olide	$C_{20}H_{30}O_3$	*C. gratissimus*	[86]
289	(+)-(1S*,4R*,8S*,10R*)-1,4,8-Trihydroxycembra-2*E*,6*E*,11*Z*-trien-20,10-olide	$C_{20}H_{30}O_5$	*C. gratissimus*	[86]
290	Cembranoid 1	$C_{20}H_{30}O_4$	*C. longissimus*	[87]
291	Cembranoid 2	$C_{20}H_{30}O_3$	*C. longissimus*	[87]

2.1.7. Abietanes

Fourteen new abietane diterpenoids (**292**–**305**) were isolated from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 7 and Table 7. Two new abietanes (**292**, **293**) were obtained from *C. megalocarpoides* [27], and *C. argyrophylloides* [63], respectively. Investigation of *C. caudatus* led to the isolation of 5 new abietanes (**294**–**298**). Among them, crotontomentosin A (**294**) was a 9,10-*seco* abietane [88]. Crotolaevigatones A–G (**299**–**305**), 7 new abietanes were found from the twigs and leaves of *C. laevigatus*, and compounds (**304**, **305**) possessed a 9,13-epidioxy moiety [89].

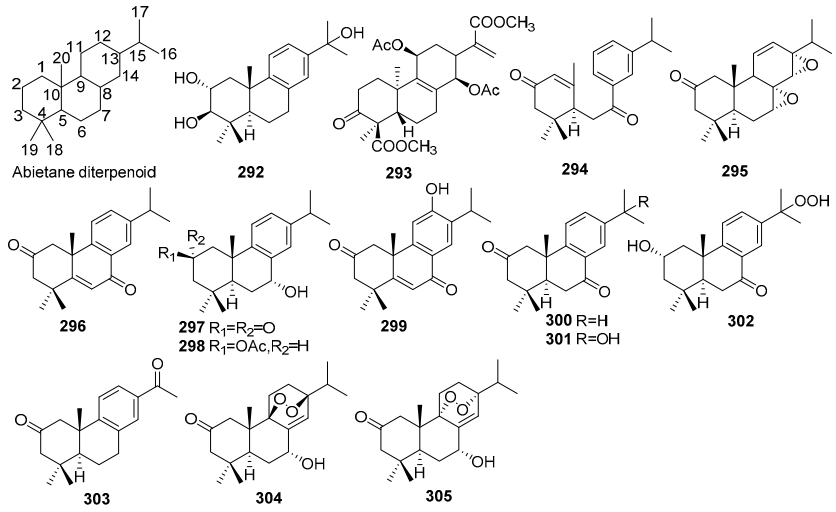

Figure 7. Abietane type diterpenoids from the genus *Croton*.

Table 7. Abietane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
292	Isolophanthin E	$C_{20}H_{30}O_3$	*C. megalocarpoides*	[27]
293	*rel*-(1*R*,4a*R*,5*R*,8*R*)-methyl-7-(1-(methoxycarbonyl)vinyl)-5,8-diacetoxy-1,2,3,4*a*,5,6,7,8,9,10,10*a*-dodecahydro-1,4*a*-dimethyl-2-oxophenanthrene-1-carboxylate	$C_{26}H_{34}O_9$	*C. argyrophylloides*	[63]
294	Crotontomentosin A	$C_{20}H_{26}O_2$	*C. caudatus*	[88]
295	Crotontomentosin B	$C_{20}H_{30}O_3$	*C. caudatus*	[88]
296	Crotontomentosin D	$C_{20}H_{24}O_2$	*C. caudatus*	[88]
297	Crotontomentosin C	$C_{20}H_{28}O_2$	*C. caudatus*	[88]
298	Crotontomentosin E	$C_{22}H_{32}O_3$	*C. caudatus*	[88]
299	Crotolaevigatone A	$C_{20}H_{24}O_3$	*C. laevigatus*	[89]
300	Crotolaevigatone B	$C_{20}H_{26}O_2$	*C. laevigatus*	[89]
301	Crotolaevigatone C	$C_{20}H_{26}O_3$	*C. laevigatus*	[89]
302	Crotolaevigatone D	$C_{20}H_{28}O_4$	*C. laevigatus*	[89]
303	Crotolaevigatone E	$C_{19}H_{24}O_2$	*C. laevigatus*	[89]
304	Crotolaevigatone F	$C_{20}H_{30}O_4$	*C. laevigatus*	[89]
305	Crotolaevigatone G	$C_{20}H_{30}O_4$	*C. laevigatus*	[89]

2.1.8. Casbanes

Seven new casbane diterpenoids (**306**–**312**) were found from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are summarized in Figure 8 and Table 8. Five new casbane s (**306**–**310**) were reported from *C. nepetaefolius* [90], and *C. argyrophyllus* [72,91], respectively. Investigations on the stem bark of *C. insularis* afforded two new casbanes, EBC-324 (**311**) and EBC-329 (**312**). Among them, EBC-329 (**312**) represented the first natural *seco*-casbane diterpene, while EBC-324 (**311**) was the first endoperoxide casbane [92].

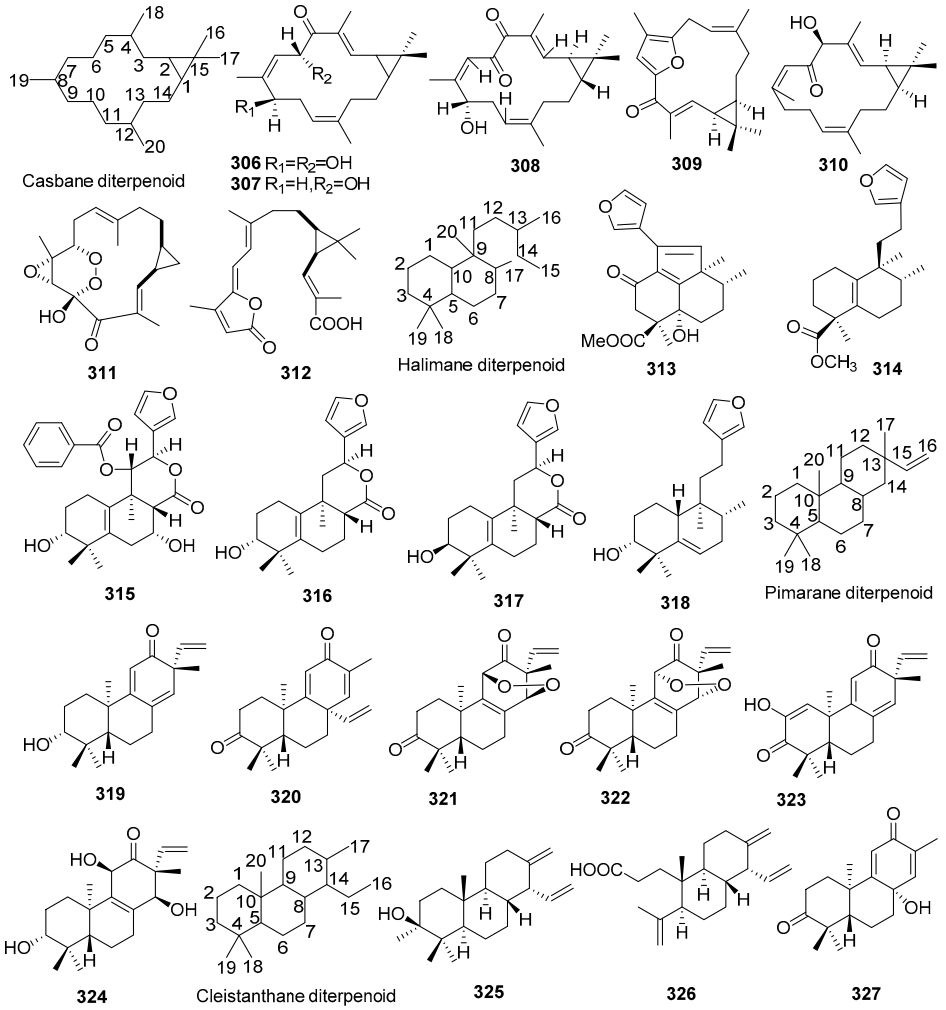

Figure 8. Casbane, Halimane, Pimarane and Cleistanthane type diterpenoids from the genus *Croton*.

Table 8. Casbane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
306	1,4-dihydroxy-2*E*,6*E*,12*E*-trien-5-one-casbane	$C_{20}H_{30}O_3$	*C.nepetaefolius*	[90]
307	4-hydroxy-2*E*,6*E*,12*E*-5-one-casbane	$C_{20}H_{29}O_3$	*C.nepetaefolius*	[90]
308	1-hydroxy-(2*E*,6*Z*,12*E*)-casba-2,6,12-triene-4,5-dione	$C_{20}H_{28}O_3$	*C.argyrophyllus*	[91]
309	6*E*,12*E*-casba-1,3,6,12-tetraen-1,4-epoxy-5-one	$C_{20}H_{26}O_2$	*C.argyrophyllus*	[91]
310	(2*E*,5*β*,6*E*,12*E*)-5-hydroxycasba-2,6,12-trien-4-one	$C_{20}H_{30}O_2$	*C. argyrophyllus*	[72]
311	EBC-324	$C_{20}H_{28}O_5$	*C. insularis*	[92]
312	EBC-329	$C_{20}H_{26}O_4$	*C. insularis*	[92]

2.1.9. Halimanes

Six new halimane diterpenoids (**313**–**318**) were reported from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 8 and Table 9. Investigations on the stem bark of *C. oblongifolius* afforded two new cleistanthanes (**325**, **326**). Among them, compound **326** was a 3,4-*seco* cleistanthane [93]. One new bis-*nor*-cleistanthane diterpenoid (**327**), was found from the twigs and leaves of *C. caudatus* [94].

Table 9. Halimane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
313	Crassifoliusin A	$C_{21}H_{24}O_5$	*C.crassifolius*	[95]
314	Crotontomentosin F	$C_{21}H_{30}O_3$	*C.caudatus*	[88]
315	Crolaevinoid A	$C_{27}H_{30}O_7$	*C.laevigatus*	[39]
316	Crolaevinoid B	$C_{20}H_{26}O_4$	*C.laevigatus*	[39]
317	Crothalimene A	$C_{20}H_{26}O_4$	*C.dichogamus*	[75]
318	Crothalimene B	$C_{20}H_{30}O_2$	*C.dichogamus*	[75]

2.1.10. Pimaranes

Six new pimarane diterpenoids (**319**–**324**) were obtained from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are listed in Figure 8 and Table 10. All six new pimaranes (**319**–**324**) were isolated from *C. insularis* [96,97]. Among them, compound **319** was an important biosynthetic intermediate.

Table 10. Pimarane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
319	*ent*-3*β*-hydroxypimara-8(14),9,15-trien-12-one	$C_{20}H_{28}O_2$	*C.insularis*	[98]
320	EBC-316	$C_{20}H_{26}O_2$	*C.insularis*	[99]
321	EBC-325	$C_{20}H_{26}O_4$	*C.insularis*	[99]
322	EBC-326	$C_{20}H_{26}O_4$	*C.insularis*	[99]
323	EBC-327	$C_{20}H_{24}O_3$	*C.insularis*	[99]
324	EBC-345	$C_{20}H_{30}O_4$	*C.insularis*	[99]

2.1.11. Cleistanthanes

Three new cleistanthane diterpenoids (**325**–**327**) were ioslated from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 8 and Table 11. Investigations on the stem bark of *C. oblongifolius* afforded two new cleistanthanes (**325**, **326**). Among them, compound **326** was a 3,4-*seco* cleistanthane [93]. One new bis-*nor*-cleistanthane diterpenoid (**327**), was found from the twigs and leaves of *C. caudatus* [94].

Table 11. Cleistanthane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
325	3-hydroxycleistantha-13(17),15-diene	$C_{20}H_{32}O$	*C.oblongifolius*	[93]
326	3,4-*seco*-cleistantha-4(18),13(17),15-trien-3-oic acid	$C_{20}H_{30}O_2$	*C.oblongifolius*	[93]
327	*rel*-(5*β*,8*α*,10*α*)-8-hydroxy-13-methylpodocarpa-9(11),13-diene-3,12-dione	$C_{18}H_{25}O_3$	*C.regelianus*	[94]

2.1.12. Grayananes, Atisanes, Phytanes, Laevinanes and Meroditerpenoids

From the leaves of *C. tonkinensis*, two new rare grayanane diterpenoids, crotonkinensins A (**328**) and B (**329**), were isolated [100]. Two new 3,4-*seco* atisane diterpenoids, crotobarin (**330**) from *C. barorum* and crotogoudin (**331**) from *C. goudotii*, were found [101]. Investigations on the aerial parts of *C. laui* gave two new phytane diterpenoids (**332**, **333**) [37]. Two new laevinane diterpenoids, crolaevinoid G (**334**) and H (**335**), were obtained [39]. Two new meroditerpenoids, steenkrotin A (**336**) and B (**337**), containing new carbon skeletons, were isolated from the leaves of *C. steenkampianus* [102]. From the the roots of *C. crassifolius*, two new meroditerpenoids, norcrassin A (**338**) and cracroson D (**339**), were reported [35,69]. Among them, norcrassin A (**338**) possessing a new carbon skeleton with a 5/5/5/6 tetracyclic system, was a C16 tetranorditerpenoid, while *c*racroson D (339) featured a new skeleton with a rare cyclobutane ring. Their structures, molecular formula, names, corresponding sources, and references are listed in Figure 9 and Tables 12–16.

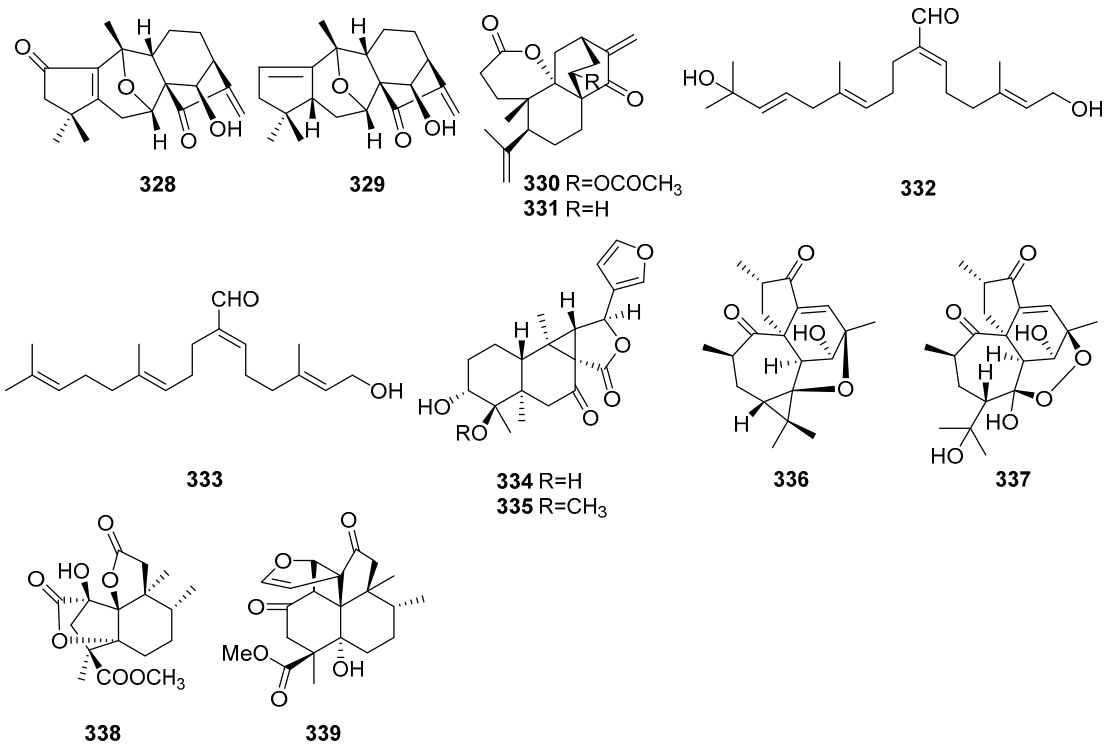

Figure 9. Grayanane, Atisane, Phytane, Laevinane type diterpenoids and Meroditerpenoids from the genus *Croton*.

Table 12. Grayanane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
328	Crotonkinensin A	$C_{20}H_{24}O_4$	*C.tonkinensis*	[100]
329	Crotonkinensin B	$C_{20}H_{26}O_3$	*C.tonkinensis*	[100]

Table 13. Atisane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
330	Crotobarin	$C_{22}H_{28}O_5$	*C.barorum*	[101]
331	Crotogoudin	$C_{20}H_{26}O_3$	*C.goudotii*	[101]

Table 14. Phytane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
332	Launine L	$C_{20}H_{32}O_3$	*C.laui*	[37]
333	Launine M	$C_{20}H_{32}O_2$	*C.laui*	[37]

Table 15. Laevinane type diterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
334	Crolaevinoid G	$C_{20}H_{24}O_6$	*C.laevigatus*	[39]
335	Crolaevinoid H	$C_{21}H_{26}O_6$	*C.laevigatus*	[39]

Table 16. Meroditerpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
336	Steenkrotin A	$C_{20}H_{24}O_6$	*C.steenkampianus*	[102]
337	Steenkrotin B	$C_{20}H_{28}O_7$	*C.steenkampianus*	[102]
338	Norcrassin A	$C_{17}H_{22}O_7$	*C.crassifolius*	[81]
339	Cracroson D	$C_{21}H_{26}O_6$	*C.crassifolius*	[40]

2.2. Sesquiterpenoids, Sesterterpenoids and Triterpenoids

Seven new sesquiterpenoids (**340**–**346**), one sesterterpenoid (**347**) and one triterpenoid (**348**) were ioslated from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are summarized in Figure 10 and Tables 17–19. From *C. muscicarpa*, one new patchoulane sesquiterpenoid (**340**) was obtained [103]. A guaiane sesquiterpenoid (**341**) was isolated from *C. regelianus* [94]. Investigations on the leaves of *C. pedicellatus* afforded a bis-*nor*-sesquiterpenoid (**342**) [104]. Two rare sesquiterpenoid, Crocrassins A (**343**) and B (**344**) having cyclopropylcyclopentane moiety, were reported [105]. Other two sesquiterpenoids, 1,3,5-cadinatriene-(7*R*,10*S*)-diol (**345**) and cracroson H (**346**) were found from *C. dichogamus* [75], and *C. crassifolius* [40], respectively. One rare sesterterpenoid, pseudopulchellol (**347**), was isolated from the leaves of *C. pseudopulchellus* [106]. From the root of *C. bonplandianum*, a new ursane triterpenoid (**348**) was obtained [107].

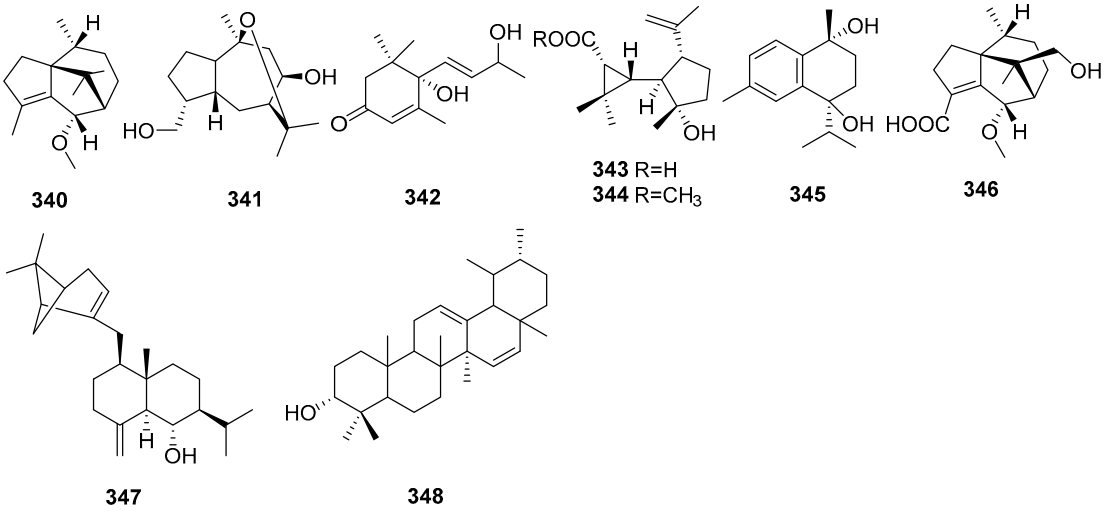

Figure 10. Sesquiterpenoids, Sesterterpenoid and Triterpenoid from the genus *Croton*.

Table 17. Sesquiterpenoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
340	6α -methoxy-cyperene	$C_{16}H_{26}O$	*C.muscicarpa*	[103]
341	*rel*-(1*R*,4*S*,6*R*,7*S*,8α*R*)-decahydro-1-(hydroxymethyl)-4,9,9-trimethyl-4,7 -(epoxymethano)azulen-6-ol	$C_{15}H_{26}O_3$	*C. regelianus*	[94]
342	Blumenol A	$C_{13}H_{20}O_3$	*C. pedicellatus*	[104]
343	Crocrassins A	$C_{15}H_{24}O_3$	*C. crassifolius*	[105]
344	Crocrassins B	$C_{16}H_{26}O_3$	*C. crassifolius*	[105]
345	1,3,5-cadinatriene-(7*R*,10*S*)-diol	$C_{15}H_{25}O_2$	*C. dichogamus*	[75]
346	Cracroson H	$C_{15}H_{22}O_3$	*C. crassifolius*	[40]

Table 18. Sesterterpenoid from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
347	Pseudopulchellol	$C_{25}H_{40}O$	*C.pseudopulchellus*	[106]

Table 19. Triterpenoid from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
348	3α-hydroxy-urs-12,15-dien	$C_{30}H_{48}O$	*C.bonplandianum*	[107]

2.3. Glycosides

Twenty-one new glycosides (**349**–**369**) were ioslated from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 11 and Table 20. From *C. crassifolius*, a patchoulane sesquiterpenoid glycoside (**349**), an isocrotofolane glucoside (**368**), and a phenolic glycoside (**369**) were reported [69,108]. Compound **350**, isolated from *C. lachnocarpus*, was the first triterpenoid glucoside reported from the genus *Croton* [109]. A new flavone glucoside (**351**) was found from the leaves of *C. zambesicus* [110]. Investigations on the leaves of *C. cascarilloides* and *C. oblongifolius* afforded 13 new megastigmane glycosides, crotonionosides A–G (**352**–**358**) and Oblongionosides A–F (**359**–**364**) [111,112]. One new bis-*nor*-sesquiterpenoid glycoside (**365**) was isolated from *C. pedicellatus* [104]. One new diglyceride galactoside (**366**) and one new clerodane glucoside (**367**) were obtained from *C. sparsiorus* [113], and *C. limae* [35], respectively.

Table 20. Glycosides from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
349	Cyperenoic acid-9-*O*-β-d-glucopyranoside	$C_{21}H_{32}O_8$	*C. crassifolius*	[108]
350	3-*O*-β-d-xylopyranosylspathodic acid	$C_{35}H_{56}O_9$	*C. lachnocarpus*	[109]
351	Helichrysoside-3′-methylether	$C_{31}H_{28}O_{14}$	*C. zambesicus*	[110]
352	Crotonionoside A	$C_{29}H_{42}O_{11}$	*C. cascarilloides*	[111]
353	Crotonionoside B	$C_{30}H_{44}O_{12}$	*C. cascarilloides*	[111]
354	Crotonionoside C	$C_{24}H_{42}O_{12}$	*C. cascarilloides*	[111]
355	Crotonionoside D	$C_{31}H_{46}O_{14}$	*C. cascarilloides*	[111]
356	Crotonionoside E	$C_{35}H_{52}O_{16}$	*C. cascarilloides*	[111]
357	Crotonionoside F	$C_{24}H_{42}O_{11}$	*C. cascarilloides*	[111]
358	Crotonionoside G	$C_{29}H_{40}O_{11}$	*C. cascarilloides*	[111]
359	Oblongionoside A	$C_{24}H_{42}O_{12}$	*C. oblongifolius*	[112]
360	Oblongionoside B	$C_{24}H_{42}O_{12}$	*C. oblongifolius*	[112]
361	Oblongionoside C	$C_{24}H_{44}O_{11}$	*C. oblongifolius*	[112]
362	Oblongionoside D	$C_{24}H_{44}O_{11}$	*C. oblongifolius*	[112]
363	Oblongionoside E	$C_{19}H_{36}O_8$	*C. oblongifolius*	[112]
364	Oblongionoside F	$C_{19}H_{36}O_8$	*C. oblongifolius*	[112]
365	Blumenol A glucoside	$C_{19}H_{30}O_8$	*C. pedicellatus*	[10]
366	Sparsioside	$C_{53}H_{102}O_{10}$	*C. sparsiorus*	[113]
367	3,12-dioxo-15,16-epoxy-4α-hydroxy-6-(β-glucopyranosyl) -ent-neo-clerodan-13(16),14-diene	$C_{26}H_{38}O_{10}$	*C. limae*	[35]
368	Isocrotofolane glucoside	$C_{26}H_{38}O_9$	*C. cascarilloides*	[69]
369	2-methoxyphenol-β-d-(6-*O*-β-d-apiofuranosyl) glucopyranoside	$C_{18}H_{26}O_{11}$	*C. cascarilloides*	[69]

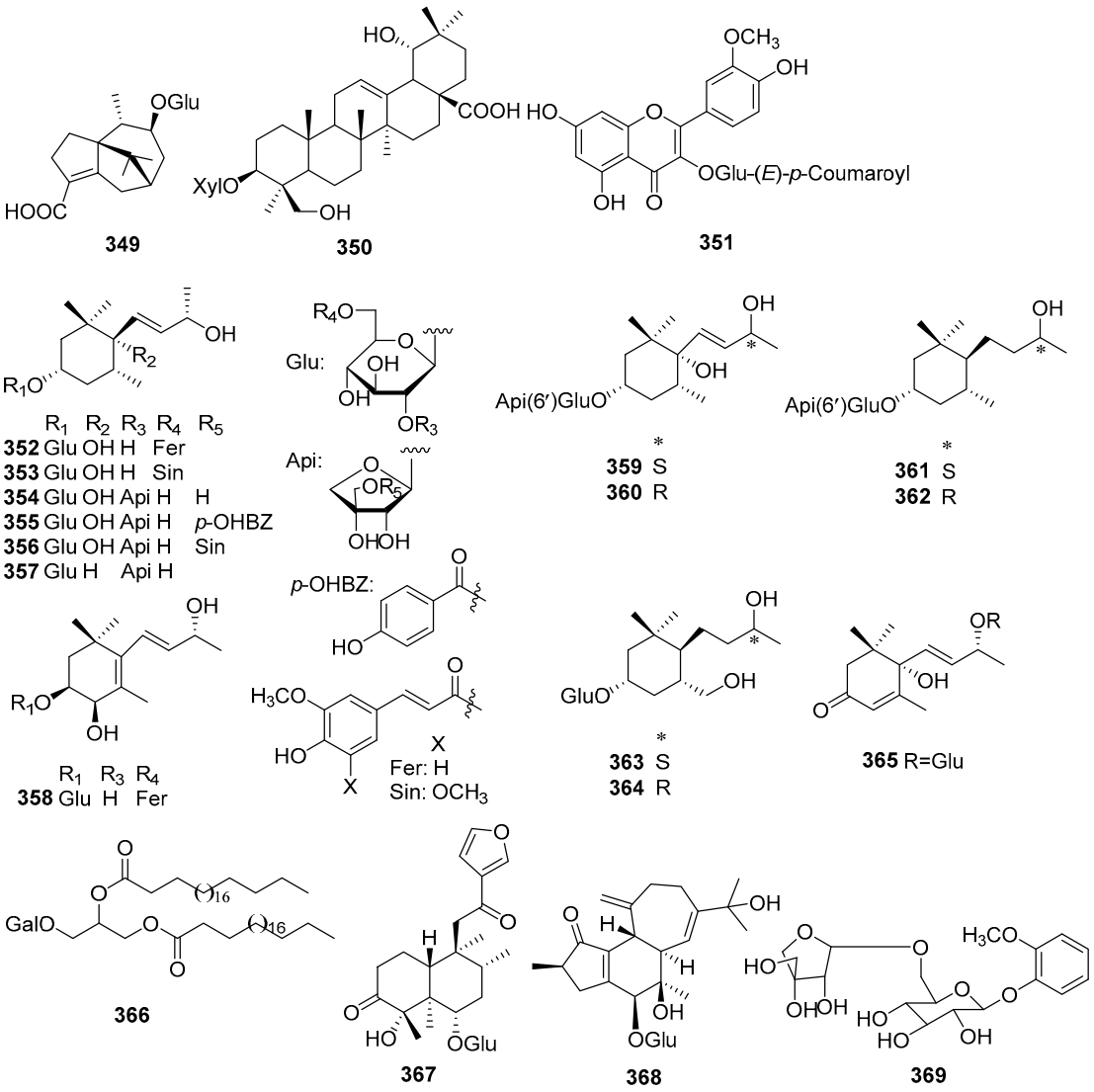

Figure 11. Glycosides from the genus *Croton*.

2.4. Alkaloids

Eight new alkaloids (**370**–**377**) were reported from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are listed in Figure 12 and Table 21. From *C. sparsiflorus*, two new amide alkaloids crotamides A (**370**) and B (**371**), and one new proaporphine alkaloid, crotsparsidine (**374**) were isolated [96,114]. One new pyrazine derivative, crotonine (**372**) was obtained from the leaves of *C. tiglium* [97]. Investigations on *C. cascarilloides* afforded a new glutarimide alkaloid, crotonimide C (**375**) [42]. Other three new alkaloids (**373**, **376**–**377**) were found from C. pullei, C. heliotropiifolius, and C. echioides, respectively [115–117].

Table 21. Alkaloids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
370	Crotamide A	$C_{36}H_{65}NO$	*C. sparsiflorus*	[114]
371	Crotamide B	$C_{38}H_{69}NO$	*C. sparsiflorus*	[114]
372	Crotonine	$C_{12}H_{14}N_2O_4$	*C. tiglium*	[97]
373	Crotonimide A	$C_{16}H_{20}N_2O_3$	*C. pullei*	[115]
374	Crotsparsidine	$C_{17}H_{17}O_3N$	*C. sparsiflorus*	[96]
375	Crotonimide C.	$C_{20}H_{20}N_2O_3$	*C. alienus*	[42]
376	6-Hydroxy-1-methyl-2-dimethyl-3,4-tetrahydro-b-carbo-line	$C_{14}H_{19}N_2O$	*C. heliotropiifolius*	[116]
377	*N-trans*-feruloyl-3,5-dihydroxyindolin-2-one	$C_{20}H_{20}N_2O_6$	*C. echioides*	[117]

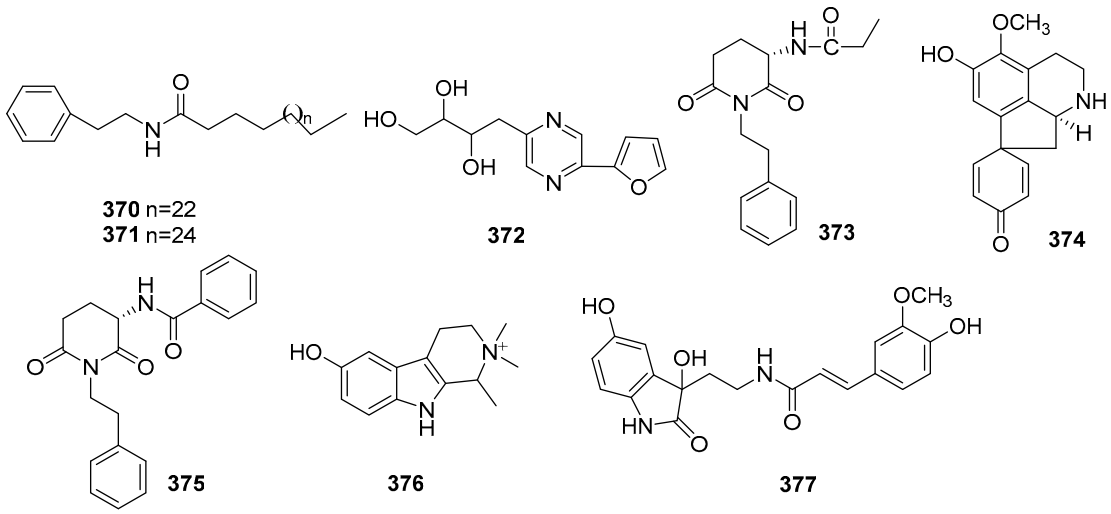

Figure 12. Alkaloids from the genus *Croton*.

2.5. Benzoate Derivatives, Pyran-2-One Derivatives, Cyclicpeptides, Tropone Derivatives and Limonoids

Three benzoate derivatives (**378**–**380**) were isolated from *C. sylvaticus* and *C. hutchinsonianus* [118,119]. Investigations on *C. crassifolius* afforded three new pyran-2-one derivatives, crotonpyrone A (**381**), B (**382**) and C (**383**) [120,121]. Two cyclicpeptides (**384**, **385**) were obtained from *C. gossypifolius* and *C. urucurana* [122,123], while two tropone derivatives (**386**, **387**) were isolated from *C. zehntneri* and *C. argyroglossum* [124,125]. From the root bark of *C. jatrophoides*, two new limonoids, musidunin (**388**) and musiduol (**389**), were found [126]. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 13 and Tables 22–26.

Table 22. Benzoate derivatives from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
378	2′-(3′′,4′′-dihydroxyphenyl)-ethyl-4-hydroxybenzoate	$C_{15}H_{14}O_5$	*C.sylvaticus*	[118]
379	3-(4-hydroxy-3,5-dimethoxyphenyl)-propyl benzoate	$C_{18}H_{20}O_5$	*C.hutchinsonianus*	[119]
380	3-(4-hydroxyphenyl)-propyl benzoate	$C_{16}H_{16}O_3$	*C.hutchinsonianus*	[119]

Table 23. Pyran-2-one derivatives from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
381	Crotonpyrone A	$C_{17}H_{28}O_3$	*C.crassifolius*	[120]
382	Crotonpyrone B	$C_{17}H_{26}O_3$	*C.crassifolius*	[120]
383	Crotonpyrone C	$C_{19}H_{28}O_3$	*C.crassifolius*	[121]

Table 24. Cyclicpeptides from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
384	Crotogossamide	$C_{37}H_{56}N_{10}O_{11}$	*C.gossypifolius*	[122]
385	[1−9-NαC]-crourorb A1	$C_{37}H_{56}N_{10}O_{11}$	*C.urucurana*	[123]

Table 25. Tropone derivatives from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
386	Crototropone	$C_{10}H_{12}O_4$	*C.zehntneri*	[124]
387	Pernambucone	$C_{15}H_{18}O_2$	*C.argyroglossum*	[125]

Table 26. Limonoids from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
388	Musidunin	$C_{31}H_{38}O_{11}$	*C.jatrophoides*	[126]
389	Musiduol	$C_{30}H_{38}O_{10}$	*C.jatrophoides*	[126]

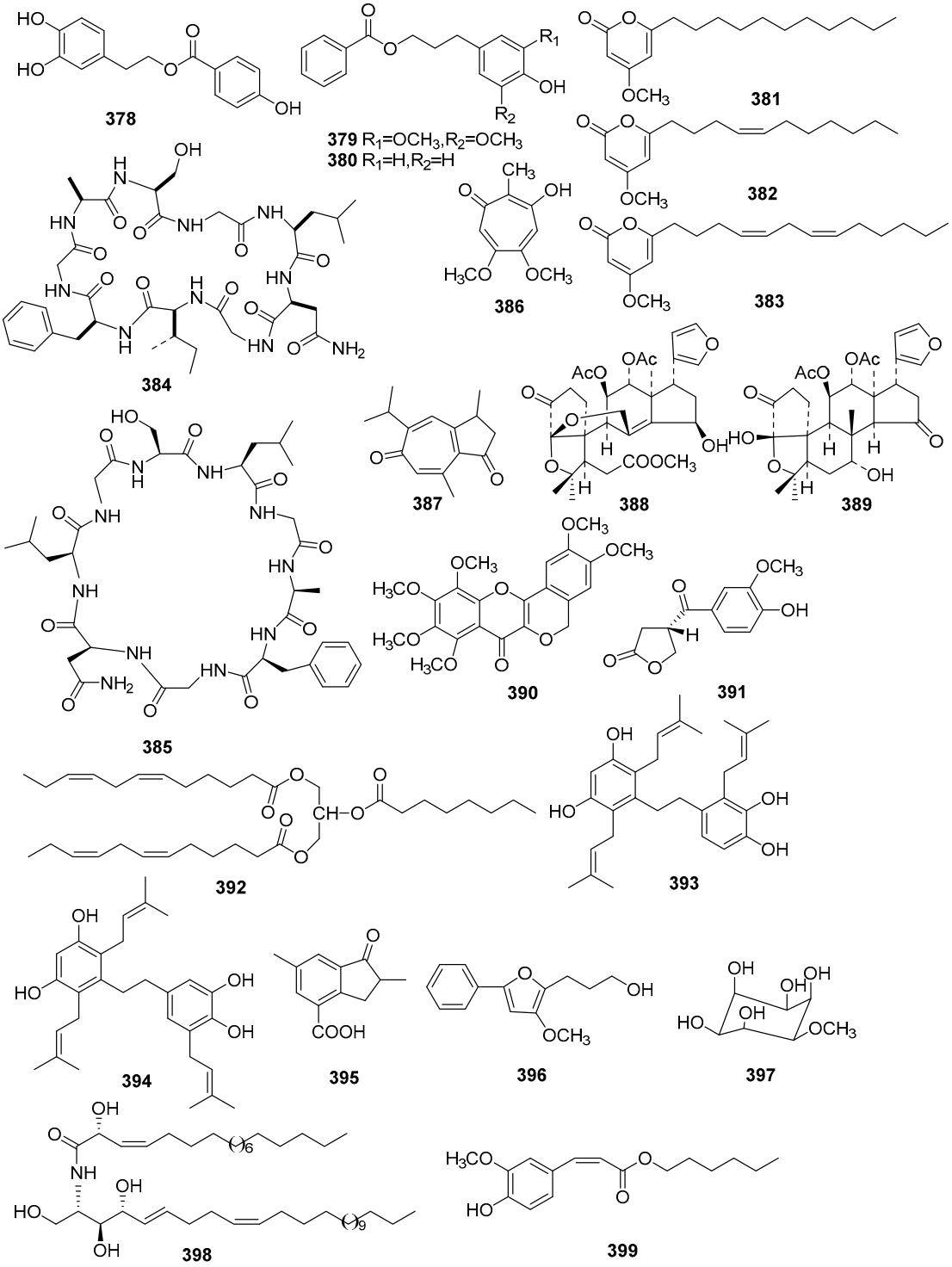

Figure 13. Miscellaneous compounds from the genus *Croton*.

2.6. Miscellaneous Compounds

Flavonoids, lignans, and other types of 10 compounds were also isolated from *Croton* species. Their structures, molecular formula, names, corresponding sources, and references are collected in Figure 13 and Table 27. From the stems of *C. caudatus*, one new flavone, crotoncaudatin (**390**), was isolated [127]. A new *nor*-lignan (**391**) was obtained from the twigs and leaves of *C. kongensis* [67]. Investigations on *C. laevifolius* gave two new prenylated dihydrostilbenes, laevifolin A (**393**), B (**394**) and one new aromatic compound (**399**) [89,128]. A long chain linear ester, lobaceride (**392**) was isolated from the twigs and leaves of *C. lobatus* [129]. One indanone derivative (**395**) was found from the leaves of *C. steenkampianus* [102], while a trisubstituted furan derivative (**396**) was isolated from the bark of *C. oblongifolius* [22]. From *C. sparsiflorus*, an inositol, sparsifol (**397**), and a sphingolipid, sparsioamide (**398**), were obtained [96,113].

Table 27. Miscellaneous compounds from the genus *Croton*.

No.	Compound Name	Molecular Formula	Sources	Ref
390	Crotoncaudatin	$C_{22}H_{22}O_9$	*C.caudatus*	[127]
391	8*S*-(−)-8-(4-hydroxy-3-methoxybenzoyl)-dihydrofuran-8(8′*H*)-one	$C_{20}H_{30}O_2$	*C.kongensis*	[67]
392	Lobaceride	$C_{35}H_{58}O_6$	*C.lobatus*	[129]
393	Laevifolin A	$C_{29}H_{38}O_4$	*C.laevifolius*	[128]
394	Laevifolin B	$C_{29}H_{38}O_4$	*C.laevifolius*	[128]
395	2,6-Dimethyl-1-oxo-4-indanecarboxylic acid	$C_{12}H_{12}O_3$	*C. steenkampianus*	[102]
396	3(3′-Methoxy-5′-phenylfuran-2′-yl)propan-1-ol	$C_{14}H_{16}O_3$	*C. oblongifolius*	[22]
397	Sparsifol	$C_7H_{15}O_6$	*C. sparsiflorus*	[96]
398	Sparsioamide	$C_{43}H_{81}NO_5$	*C. sparsiflorus*	[113]
399	hexyl Z-ferulate	$C_{16}H_{22}O_4$	*C. laevigatus*	[89]

3. Biological Activities

Compounds isolated from *Croton* species exert a wide range of biological activities, including cytotoxic, anti-inflammatory, antifungal, acetylcholinesterase inhibitory, and neurite outgrowth-promoting activities.

3.1. Cytotoxic Activity

The anti-tumor activity of many plants from the *Croton* species have been reported. Therefore, the cytotoxicity of the isolated compounds is the most commonly studied bioactivity. The cytotoxic activities of the isolated compounds from the *Croton* species are listed in Table 28. Four new tigliane diterpene esters (**135**–**137**, **139**) from the leaves of *C. tiglium*, exhibited most potent cytotoxic activity against K562 cell line with IC_{50} values of 0.03, 0.03, 0.07 and 0.05 μM, respectively [51].

Table 28. Cytotoxic activity of compounds from the genus *Croton*.

Compounds	Tumor Cell Line	Activity (IC_{50})	Ref
Methyl 15,16-epoxy-3,13(16),14-ent-clerodatrien-18,19-olide-17-carboxylate (**6**)	HuCCA-1	36.0 μg/mL	[29]
	KB	26.0 μg/mL	[29]
	HeLa	30.0 μg/mL	[29]
	MDA-MB231	29.0 μg/mL	[29]
	T47D	10.0 μg/mL	[29]
Dimethyl-15,16-epoxy-12-oxo-3,13 (16)14-ent-clerodatriene-17,18-dicarboxylate (**7**)	HuCCA-1	39.0 μg/mL	[29]
	KB	27.0 μg/mL	[29]
	HeLa	29.0 μg/mL	[29]
	MDA-MB231	27.0 μg/mL	[29]
	T47D	25.0 μg/mL	[29]
Laevigatbenzoate (**8**)	HeLa	45.4 μM	[13]
Crotonolide A (**23**)	HL-60	9.42 μM	[21]
	P-388	7.45 μM	[21]
15-oxo-17(10′-α-pinenyl)-kauran-18-oic acid (**181**)	HCT-116	7.14 μg/mL	[35]
	OVCAR-8	8.19 μg/mL	[35]
	SF-295	>10.0 μg/mL	[35]
Launine K (**67**)	HeLa	14.5 μM	[37]
	MCF-7	62.5 μM	[37]
Crassin H (**75**)	HL-60	11.8 ± 2.1 μM	[17]
	A549	5.2 ± 0.4 μM	[17]

Table 28. *Cont.*

Compounds	Tumor Cell Line	Activity (IC_{50})	Ref
Crassifolius A (**76**)	Hep3B	17.91 μM	[38]
	HepG2	42.04 μM	[38]
Cracroson D (**339**)	T24	14.48 ± 0.65 μM	[40]
	A549	25.64 ± 2.14 μM	[40]
Cracroson E (**87**)	T24	22.99 ± 1.76 μM	[40]
	A549	51.88 ± 14.07μM	[40]
12-*O*-benzoylphorbol-13-(2-methyl)butyrate (**114**)	Hela	3.9 μM	[48]
	DU145	7.2 μM	[48]
	A549	5.8 μM	[48]
	SGC-7091	13 μM	[48]
	H1975	10 μM	[48]
	HL60	12 μM	[48]
	293T	291.6 μM	[48]
	LX-2	>500.0 μM	[48]
	K562	15 μM	[48]
	MOLT-4	12 μM	[48]
	U937	17 μM	[48]
	MCF-7	20 μM	[48]
12-*O*-tiglyl-7-oxo-5-ene-phorbol-13-(2-methyl)butyrate (**115**)	Hela	4.6 μM	[48]
	DU145	4.3 μM	[48]
	A549	6.9 μM	[48]
	SGC-7091	10 μM	[48]
	H1975	3.3 μM	[48]
	HL60	6.8 μM	[48]
	293T	420.4 μM	[48]
	LX-2	>500.0 μM	[48]
	K562	17 μM	[48]
	MOLT-4	4.8 μM	[48]
	U937	21 μM	[48]
	MCF-7	20 μM	[48]
13-*O*-(2-metyl)butyryl-4-deoxy-4a-phorbol (**116**)	Hela	5.0 μM	[48]
	DU145	10 μM	[48]
	A549	19 μM	[48]
	SGC-7091	23 μM	[48]
	H1975	10 μM	[48]
	HL60	10 μM	[48]
	293T	455.3 μM	[48]
	LX-2	>500.0 μM	[48]
	K562	8.0 μM	[48]
	MOLT-4	9.9 μM	[48]
	U937	18 μM	[48]
	MCF-7	24 μM	[48]
	H1975	10 μM	[48]
	HL60	10 μM	[48]
	293T	455.3 μM	[48]
	LX-2	>500.0 μM	[48]
	Hela	10 μM	[48]
	DU145	10 μM	[48]
	A549	4.5 μM	[48]
	SGC-7091	5.4 μM	[48]
	H1975	3.3 μM	[48]
	HL60	9.8 μM	[48]
	293T	191.0 μM	[48]
	LX-2	>500.0 μM	[48]
Crotignoid A (**117**)	HL-60	1.61 μM	[49]
	A549	2.85 μM	[49]
Crotignoid B (**118**)	HL-60	22.1 μM	[49]
	A549	31.0 μM	[49]
Crotignoid C (**119**)	HL-60	32.3 μM	[49]
	A549	5.03 μM	[49]
Crotignoid D (**120**)	HL-60	19.8 μM	[49]
	A549	10.2 μM	[49]
Crotignoid F (**122**)	HL-60	44.6 μM	[49]
	A549	6.96 μM	[49]
Crotignoid G (**123**)	HL-60	22.1 μM	[49]
	A549	3.89 μM	[49]
Crotignoid H (**124**)	HL-60	9.97 μM	[49]
	A549	8.08 μM	[49]
Crotignoid I (**125**)	HL-60	14.8 μM	[49]
	A549	24.4 μM	[49]
Crotignoid J (**126**)	HL-60	14.2 μM	[49]
	A549	29.5 μM	[49]
Crotusin A (**128**)	HL-60	12.53 ± 0.37 μM	[44]
	SMMC-7721	7.06 ± 0.72 μM	[44]
	A549	9.69 ± 0.41 μM	[44]
	MCF-7	9.56 ± 0.76 μM	[44]
	SW480	14.88 ± 0.43 μM	[44]
Crotusin B (**129**)	HL-60	19.39 ± 0.46 μM	[44]
	SMMC-7721	21.13 ± 0.29 μM	[44]
	A549	14.66 ± 1.66 μM	[44]
	MCF-7	1.49 ± 0.23 μM	[44]
	SW480	31.21 ± 3.20 μM	[44]
Crotusin C (**130**)	HL-60	4.19 ± 0.15 μM	[44]

Table 28. *Cont.*

Compounds	Tumor Cell Line	Activity (IC_{50})	Ref
	SMMC-7721	3.87 ± 0.12 μM	[44]
	A549	2.44 ± 0.35 μM	[44]
	MCF-7	0.49 ± 0.04 μM	[44]
	SW480	2.89 ± 0.01 μM	[44]
12-*O*-tiglylphorbol-4-deoxy-4*β*-phorbol-13-acetate (**131**)	SNU387	59.5 ± 2.1 μM	[50]
	SNU398	43.7 ± 1.5 μM	[50]
12-*O*-tiglylphorbol-4-deoxy-4*β*-phorbol-13-hexadecanoate (**132**)	SNU387	30.2 ± 1.4 μM	[50]
	SNU398	91.2 ± 3.7 μM	[50]
13-*O*-acetylphorbol-4-deoxy-4*β*-phorbol-20-oleate (**133**)	SNU387	1.9 ± 0.2 μM	[50]
	SNU398	13.5 ± 1.1 μM	[50]
13-*O*-acetylphorbol-4-deoxy-4*β*-phorbol-20-linoleate (**134**)	SNU387	0.71 ± 0.08 μM	[50]
	SNU398	18.2 ± 1.7 μM	[50]
4-deoxy-20-oxophorbol 12-tiglyl 13-acetate (**135**)	K562	0.03 μM	[51]
	A549	6.88 μM	[51]
	Huh-7	3.85 μM	[51]
7-oxo-5-ene-phorbol-13-(2-methylbutyrate) (**136**)	K562	0.03 μM	[51]
	A549	6.33 μM	[51]
	Huh-7	20.9 μM	[51]
7-hydroxyl-phorbol-5-ene-13-(2-methyl)butyrate (**137**)	K562	0.07 μM	[51]
	A549	8.86 μM	[51]
	Huh-7	11.6 μM	[51]
13-*O*-(2-metyl)butyryl-phorbol (**139**)	K562	0.05 μM	[51]
	A549	43.5 μM	[51]
	Huh-7	34.2 μM	[51]
7-keto-12-*O*-tiglylphorbol-13-acetate (**140**)	HL-60	6.22 ± 3.24 μg/mL	[52]
	A549	18.0 ± 9.48 μg/mL	[52]
Phorbol-13-isobutyrate (**148**)	HL-60	0.22 ± 0.15 μg/mL	[52]
14-*epi*-hyalic acid (**159**)	HL-60	8.2 μM	[63]
Kongeniod A (**178**)	HL-60	1.27 ± 0.24 μM	[59]]
	A549	5.74 ± 0.25 μM	[59]
Kongeniod B (**179**)	HL-60	0.47 ± 0.04 μM	[59]
	A549	3.25 ± 0.91 μM	[59]
Kongeniod C (**180**)	HL-60	0.58 ± 0.17 μM	[59]
Crotonkinensin D (**188**)	MCF-7	9.4 ± 1.7 μM	[61]
	MCF-7/TAMR	2.6 ± 0.9 μM	[61]
	MCF-7/ADR	18.9 ± 0.6 μM	[61]
	MDA-MB-231	22.0 ± 0.9 μM	[61]
EBC-162 (**207**)	HL-60	15 μg/mL	[74]
	HT29	15 μg/mL	[74]
	MCF-7	30 μg/mL	[74]
	MM96	10 μg/mL	[74]
	NNF	20 μg/mL	[74]
	K562	50 μg/mL	[74]
EBC-233 (**208**)	HL-60	10 μg/mL	[74]
	HT29	80 μg/mL	[74]
	MCF-7	20 μg/mL	[74]
	MM96	6 μg/mL	[74]
	NNF	50 μg/mL	[74]
	K562	50 μg/mL	[74]
EBC-300 (**209**)	HL-60	35 μg/mL	[74]
	HT29	100 μg/mL	[74]
	MCF-7	100 μg/mL	[74]
	MM96	80 μg/mL	[74]
	NNF	80 μg/mL	[74]
	K562	100 μg/mL	[74]
EBC-240 (**210**)	HL-60	45 μg/mL	[74]
	HT29	80 μg/mL	[74]
	MCF-7	50 μg/mL	[74]
	MM96	12 μg/mL	[74]
	NNF	80 μg/mL	[74]
	K562	60 μg/mL	[74]
EBC-241 (**211**)	HL-60	40 μg/mL	[74]
	HT29	80 μg/mL	[74]
	MCF-7	40 μg/mL	[74]
	MM96	12 μg/mL	[74]
	NNF	75 μg/mL	[74]
	K562	60 μg/mL	[74]
Furanocembranoid 1 (**266**)	BT474	7.8 μg/mL	[83]
	CHAGO	7.0 μg/mL	[83]
	Hep-G2	5.6 μg/mL	[83]
	KATO-3	5.9 μg/mL	[83]
	SW-620	6.3 μg/mL	[83]
Furanocembranoid 2 (**267**)	BT474	9.5 μg/mL	[83]
	CHAGO	>10 μg/mL	[83]
	Hep-G2	>10 μg/mL	[83]
	KATO-3	6.8 μg/mL	[83]
	SW-620	9.9 μg/mL	[83]
Furanocembranoid 3 (**268**)	BT474	9.6 μg/mL	[83]
	CHAGO	7.1 μg/mL	[83]
	Hep-G2	5.7 μg/mL	[83]
	KATO-3	8.2 μg/mL	[83]
	SW-620	5.6 μg/mL	[83]]

Table 28. *Cont.*

Compounds	Tumor Cell Line	Activity (IC_{50})	Ref
Furanocembranoid 4 (**269**)	BT474	9.6 μg/mL	[83]
	CHAGO	9.3 μg/mL	[83]
	Hep-G2	6.1 μg/mL	[83]
	KATO-3	8.1 μg/mL	[83]
	SW-620	6.0 μg/mL	[83]
Laevigatlactone B (**272**)	Hela	38.4 μM	[84]
(+)-[1R*,2S*,7S*,8S*,12R*]-7,8-Epoxy-2,12-cyclocembra-3*E*,10*Z*dien-20,10-olide (**276**)	PEO1	132 nM	[85]
	PEO1TaxR	200 nM	[85]
(+)-[1R*,4S*,10R*]-4-Hydroxycembra-2*E*,7*E*,11*Z*-trien-20,10-olide (**278**)	PEO1	125 nM	[85]
	PEO1TaxR	135 nM	[85]
Crotontomentosin A (**294**)	Hela	24.0 ± 2.6 μM	[88]
	Hep G2	87.9 ± 4.5 μM	[88]
	MDA-MB-231	54.1 ± 2.1 μM	[88]
	A549	40.6 ± 3.9 μM	[88]
Crotontomentosin B (**295**)	Hela	>100 μM	[88]
	Hep G2	28.1 ± 2.1 μM	[88]
	MDA-MB-231	28.7 ± 3.4 μM	[88]
	A549	29.1 ± 5.2 μM	[88]
Crotontomentosin C (**297**)	Hela	47.9 ± 3.3 μM	[88]
	Hep G2	83.3 ± 5.3 μM	[88]
	MDA-MB-231	>100 μM	[88]
	A549	>100 μM	[88]]
Crotontomentosin D (**296**)	Hela	59.7 ± 4.5 μM	[88]
	Hep G2	>100 μM	[88]
	MDA-MB-231	49.3 ± 2.8 μM	[88]
	A549	>100 μM	[88]
Crotolaevigatone B (**300**)	A549	21.2 μM	[89]
	MDA-MB-231	33.4 μM	[89]
Crotolaevigatone G (**305**)	A549	25.6 μM	[89]
	MDA-MB-231	32.7 μM	[89]
EBC-324 (**311**)	MCF-7	40 μM	[92]
	NFF	50 μM	[92]
	K562	6 μM	[92]
EBC-329 (**312**)	MCF-7	13 μM	[92]
	NFF	40 μM	[92]
	K562	0.6 μM	[92]
ent-3*β*-hydroxypimara-8(14),9,15-trien-12-one (**319**)	NFF	23 μg/mL	[98]
	Hela	13 μg/mL	[98]
	HT 29	13 μg/mL	[98]
	MCF-7	16 μg/mL	[98]
	MM96L	2.8 μg/mL	[98]
	K562	17 μg/mL	[98]
EBC-325 (**321**)	MCF-7	20 μM	[99]
	NFF	6 μM	[99]
	K562	3 μM	[99]
EBC-326 (**322**)	MCF-7	14 μM	[99]
	NFF	6 μM	[99]
	K562	6 μM	[99]
EBC-327 (**323**)	MCF-7	10 μM	[99]
	NFF	10 μM	[99]
	K562	10 μM	[99]
3-hydroxycleistantha-13(17),15-diene (**325**)	KATO-3	6.0 μg/mL	[93]
	SW-620	>10 μg/mL	[93]
	BT474	6.1 μg/mL	[93]
	Hep-G2	0.5 μg/mL	[93]
	CHAGO	5.5 μg/mL	[93]
3,4-*seco*-cleistantha-4(18),13(17),15-trien-3-oic acid (**326**)	KATO-3	9.6 μg/mL	[93]
	SW-620	>10 μg/mL	[93]
	BT474	10 μg/mL	[93]
	Hep-G2	8.6 μg/mL	[93]
	CHAGO	>10 μg/mL	[93]
Crotobarin (**330**)	KB	2.5 ± 0.10 μM	[101]
	HT29	2.1 ± 0.60 μM	[101]
	A549	0.79 ± 0.15 μM	[101]
	HL60	0.56 ± 0.02 μM	[101]
Crotogoudin (**331**)	KB	1.5 ± 0.03 μM	[101]
	HT29	1.9 ± 0.25 μM	[101]
	A549	0.54 ± 0.02 μM	[101]
	HL60	0.49 ± 0.01 μM	[101]
Crotonpyrone A (**381**)	Hela	10.21 μg/mL	[120]
	NCI-446	6.59 μg/mL	[120]
Crotonpyrone B (**382**)	Hela	9.54 μg/mL	[120]
[1−9-N*α*C]-crourorb A1 (**385**)	NCI-ADR/RES	4.8 μM	[123]

3.2. Anti-Inflammatory Activity

Bioassay-guided fractionation of the aerial parts of *C. ciliatoglandulifer* led to the isolation of tigliane diterpenoids **95**, **97**, which inhibited the enzymes cyclooxygenases-1 (IC_{50}, 0.001, and 1.0 μM, respectively) and cyclooxygenases-2 (IC_{50}, 2.2 μM, for compound **95**) [41]. A tigliane diterpenoid (**114**) was isolated from the branches and leaves of C. *tiglium*, which displayed moderate inhibition of the

enzymes COX-1 and COX-2, with IC_{50} values of 0.14 and 8.5 μM, respectively [48]. crotonkinin A (**157**), isolated from *C. tonkinensis*, showed anti-inflammatory effect on LPS-induced iNOS-dependent NO production and NOX-dependent ROS production in microglial cells (IC_{50}, 46.2 ± 3.1 μM in NOS; maximum inhibition of NOX activity at 50 μM, 11.2%) [62]. Eight *ent*-kauranes (**169**–**176**) from *C. tonkinensis* exhibited the anti-inflammatory potential for inhibition of superoxide Anion generation and elastase release. Among them, crotonkinins F (**172**) displayed significant inhibition of superoxide anion generation (IC_{50}, 2.88 ± 0.52 μM) and elastase release (IC_{50}, 4.44 ± 1.45 μM) [66]. Labdane diterpenoids **251**, **254** and **257**, **258**, isolated from the aerial parts of *C. laui*, were found to show anti-inflammatory activities in LPS-stimulated RAW 264.7 cells with IC_{50} values in the range 42.73–93.04 μM [82]. Two grayanane diterpenoids, crotonkinensins A (**328**) and B (**329**) from the leaves of *C. tonkinensis*, were reported to decrease the LPS-induced COX-2 promoter activity in Raw 264.7 cells with IC_{50} values of 7.14 ± 0.2 and 5.49 ± 0.2 μM, respectively [100]. Two benzoate derivatives (**379**, **380**) were obtained from *C. hutchinsonianus*. Compound **379** showed significant activity against COX-1 (IC_{50}, 4.95 ± 0.58 μg/mL) and COX-2 (IC_{50}, 2.11 ± 1.3 μg/mL), while compound **380** (IC_{50}, 1.88 ± 0.17 μg/mL) preferentially inhibited COX-2 [119].

3.3. Antifungal Activity

Two benzoate derivatives (**379**–**380**) were isolated from *C. hutchinsonianus*, and exhibited antifungal activity against *Candida albicans* (IC_{50}, 11.41 ± 1.44, and 5.36 ± 0.01 μg/mL, respectively) [119]. Ursane triterpenoid (**348**) from the root of *C. bonplandianum*, displayed the antifungal activity against *Calletotricheme camellie* (IC_{50}, 10 μg/mL), *Fussarium equisitae* (IC_{50}, <15 μg/mL), *Alterneria alternata* (IC_{50}, 10 μg/mL), *Curvularia eragrostidies* (IC_{50}, <10 μg/mL) and *Colletorichum gloeosporiodes* (IC_{50}, 15 μg/mL) [107].

3.4. Acetylcholinesterase Inhibitory Activity

An indole alkaloid derivative **376**, isolated from the leaves of *C. heliotropiifolius*, exhibited the acetylcholinesterase inhibitory activity with IC_{50} values of 17.8 ± 0.6 μM [116]. Compund **378** from *C. sylvaticus*, also displayed the same activity [118].

3.5. Neurite Outgrowth-Promoting Activity

Two clerodane diterpenoids, crotonpenes A (**36**) and B (**37**) were isolated from *C. yanhuii*, which markedly increased the NGF (20 ng/mL)-induced proportion of neurite bearing cells by 59%, and 47% at 15 μM, respectively [23]. Crotoeurins A–C (40–42) obtained from *C. euryphyllus*, were found to display neurite outgrowth-promoting activity on NGF mediated PC12 cells at concentration of 10 μM. The percentages of neurite-bearing cells were 9.72%, 14.90%, and 7.14%, respectively [25].

3.6. Other Activities

Besides the above activities, other biological activities have also been reported. Crotonolide G (**32**), from the aerial parts of *C. laui*, was found to exhibit potent antibacterial activity (MIC, 43.4 μM) against four strains of Gram-positive bacteria, namely, *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Micrococcus luteus*, and *Bacillus subtilis* [21]. Crassifolin H (**39**) was obtained from roots of *C. crassifolius* as an angiogenic inhibitor by reducing vessel formation to 59.3% at 15 μg/mL [34]. Tigliane diterpene (**111**) was isolated from the leaves of *C. mauritianus*, which inhibited chikungunya virus-induced cell death in cell culture with EC_{50}s of 4.0 ± 0.8 μM [43]. The leaves of *C. tiglium* yielded two tigliane diterpenoids (**135**, **136**), which displayed significant antitubercular activities with MIC values of 19.5, and 20.9 μM, respectively [51]. Compounds (**162**–**165**) were four ent-kaurane diterpenes from *C. tonkinensis*, which significantly stimulated differentiation in osteoblasts [64]. From the twigs and leaves of *C. cascarilloides*, two crotofolane diterpenoid alkaloids cascarinoids B–C (**226**, **227**) were obtained, both of which displayed moderate activities against the ConA-induced proliferation of T lymphocyte cells and/or LPS-induced proliferation of B lymphocyte cells with IC_{50} values ranging

from 6.14 to 16.27 μM [71]. Meroditerpenoid (336), from *C. steenkampianus*, showed antiplasmodial activities of 15.8 (D10), 9.1 (W2), and 9.4 (Dd2) μM [102]. Indole alkaloid (**377**) was found in *C. mauritianus* with antioxidant activity (IC_{50}, 30.0.0 ± 0.7 μM) by the DPPH radical scavenging assay [117]. Bioactivity-guided fractionation of the root bark of *C. jatrophoides* resulted in the isolation of musidunin (**388**) and musiduol (**389**), both of which showed insect antifeedant activities (PC_{50}, 3 μg/mL, PC_{95}, 10 μg/mL; PC_{50}, 4 μg/mL, PC_{95}, 20 μg/mL, respectively) against the second-instar larvae of *Pectinophora gossypiella* in a leaf disk assay [126].

4. Conclusions

In the present review, we systematically summarized the chemical constituents and biological activity studies of *Croton* species covering from 2006 to 2018. To date, a total of 399 new compounds were reported from *Croton* species, which included 339 diterpenoids, seven sesquiterpenoids, 21 glycosides, eight alkaloids, and 24 miscellaneous compounds (Figure 14). Obviously, diterpenoids are characteristic components for *Croton* species. The diterpenoids with clerodane, tigliane, kaurane, crotofolane, labdane, and cembrane skeletons are among the most studied diterpenoids isolated from *Croton* species (Figure 14). Although the current studies have shown that these isolated compounds from *Croton* species possessed diversified biological activities, many compounds have never been biologically tested. Moreover, most studies conducted so far have focused mainly on in vitro cytotoxic assays. Further studies on the mechanism of actions and the structure activity relationship are needed in order to provide a better understanding of the chemical constituents from *Croton* species as potential medicines. Increasing interest in the chemistry and pharmaceutics of *Croton* species may promote new progress in finding and developing novel compounds.

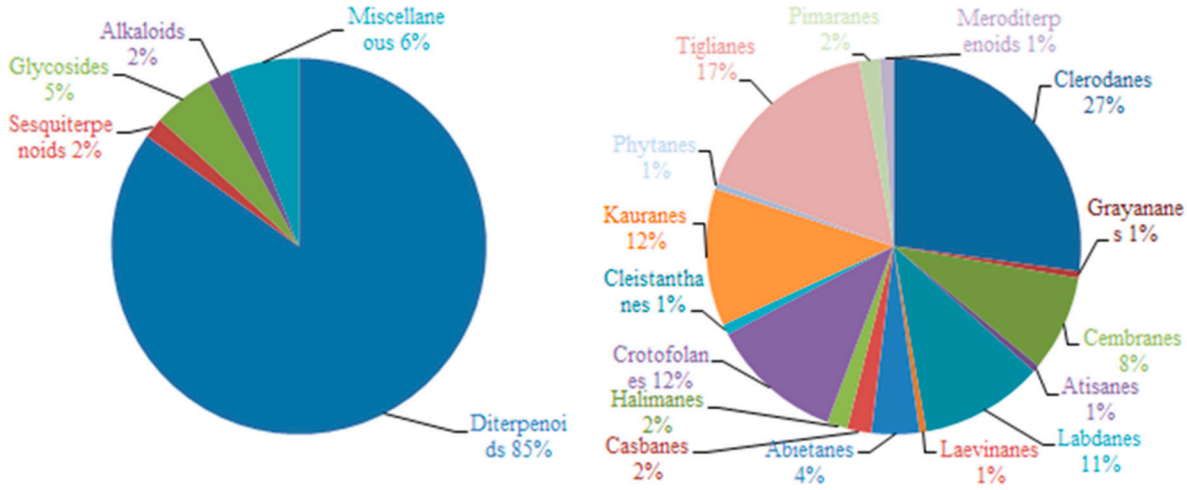

Figure 14. The percentage of each type of compounds (**left**), the percentage of each type of diterpenoids (**right**) from *Croton* Species.

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