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REVIEW

THE SIMPLE β-CARBOLINE ALKALOIDS

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Key Word Index—Spermatophyta; alkaloids; β-carbolines; pyrido(3, 4-b)indole; harman; review.

Abstract—The simple β-carbolines are presented as a distinct group of alkaloids displaying structural modification of three basic ring systems. Their occurrence in plants is listed on a taxonomic basis and aspects of their metabolism, pharmacological properties and taxonomic significance are briefly summarized.

The indole alkaloids comprise a group of widespread natural occurrence, displaying considerable structural diversity. Many of the indole alkaloids contain an intact β-carboline skeleton. This review will be confined to discussion of the 'simple' β-carbolines which are here defined as compounds comprising the tricyclic pyrido(3, 4-b)indole ring system [1] with alkyl C_1 substituents. This definition therefore excludes compounds containing additional fused-ring systems or those with cyclic C_1 substituents, the most common of which are of terpenoid origin and are more closely related to the tetra- and penta-cyclic β-carboline alkaloids [3, 24]. The simple β-carbolines, under various definitions, have been the subject of previous specialist reviews [4–8] in addition to being covered in standard texts on the alkaloids. The closely related indolalkylamines have been the subject of recent review [9]. This review covers reports in major journals and *Chemical Abstracts* to April 1979.

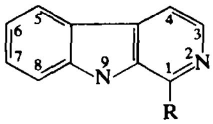
The simple β-carbolines present a variety of modifications, by simple radical substitutions, of three basic ring systems characterized by the hydrogenation state of the pyridyl ring (Table 1, **A–C**: numbering convention that of *Chemical Abstracts* [1]). The most commonly used trivial nomenclature is based on the root —harm—, derived from an early isolation from *Peganum harmala* L. [226], although a number of alternative trivial names have proposed independently (Table 1). The generic term 'tryptoline' has been proposed for the 1,2,3,4-tetrahydro-β-carbolines (Table 1: ring **C**), following reports of their formation and endogenous occurrence in animal tissues [10].

Organic synthesis of β-carbolines has been investigated extensively [4, 11]. The 1,2,3,4-tetrahydro-β-carbolines can be formed by cyclization of the Schiff base intermediates derived from the condensation of tryptophan and tryptamine with aldehydes, and of tryptamine with α-keto acids. These reactions have been shown to take place under quasi-physiological conditions of temperature, concentration and pH (see [4]), thus suggesting their occurrence *in vivo*. A subsequent stepwise oxidation has then been proposed to take place, giving rise to the 3,4-dihydro-β-carbolines and β-carbolines [12]. Alternative biogenetic pathways have been proposed for the derivation of 1,2,3,4-tetrahydro-β-carbolines [13] and for the direct formation of 3,4-dihydro-β-carbolines and β-carbolines, without participation of the 1,2,3,4-tetrahydro forms as intermediates [14, 15].

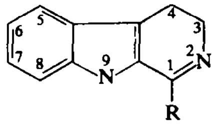
The indolyl moiety of the tricyclic ring system has been demonstrated to be derived by biosynthesis from tryptophan by using isotope-labelled precursors in feeding experiments with *Elaeagnus angustifolia* [16, 17], *Passiflora edulis* [18], *Carex brevicollis* [19], and *Peganum harmala* [20–23]. Tryptophan is also a major precursor for the indolyl moiety of the more complex indole alkaloids containing a β-carboline skeleton [2, 3]. These studies have provided evidence for the biosynthesis of these alkaloids from an amino acid precursor thus verifying a biogenetic concept which is widely accepted to be of general application to alkaloid biosynthesis [25].

In contrast, the origin of the 'non-tryptophan' biosynthetic condensation adducts and the intermediates in the metabolism of the β-carbolines *in vivo* are not so clear. Incorporation of pyruvate-[^{14}C] and acetate-[^{14}C] into the pyridyl ring of simple β-carbolines has been demonstrated in *Elaeagnus angustifolia* [16, 17] and *Peganum harmala* [21], and was presumed to take place via Schiff base intermediates. An alternative biosynthetic pathway has been proposed through *N*-acetylation of tryptamine and subsequent cyclo-dehydration to the 3,4-dihydro-β-carbolines, based on evidence from feeding and trapping experiments in *Passiflora edulis* [18]. However, the subsequently demonstrated inoperation of this pathway in *Elaeagnus angustifolia* [17, 26] and the apparent scarcity of the naturally occurring 3,4-dihydro forms (Table 2) suggests that this is not the only pathway for the biosynthesis of these alkaloids in

Table 1. Structures of simple β-carboline alkaloids



A



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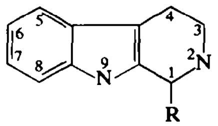


C

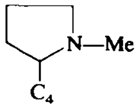
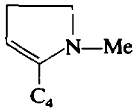
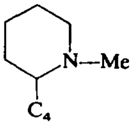
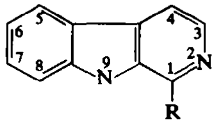
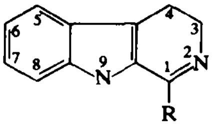
No.	Trivial name	Ring system	R	Additional substituents and their location
1.	Norharman	A	H	—
2.	—	A	H	N_2—Me; C_6—OMe
3.	—	A	H	C_7—COOMe
4.	Norharmalan	B	H	—
5.	Tetrahydronorharman Noreleagnine	C	H	—
6.	—	C	H	N_2—Me
7.	—	C	H	N_2—Me; C_6—OMe
8.	—	C	H	N_2—Me; C_6—OMe; N_9—Me
9.	Plectocomine	C	H	C_6—OH
10.	—	C	H	C_6—OMe
11.	Harman Passiflorine Locuturine Loturine Aribine Zygofabagin	A	Me	—
12.	Harmanine	A	Me	N_2—OH
13.	Melinonine F	A	Me	N_2—Me
14.	—	A	Me	N_2—O; C_7—OMe
15.	—	A	Me	C_3—COOH
16.	—	A	Me	C_3—COOMe
17.	Brevicarine	A	Me	C_4—$(CH_2)_4$NHMe
18.	Brevicolline	A	Me	
19.	Dehydrobrevicolline	A	Me	
20.	Homobrevicolline	A	Me	
21.	—	A	Me	C_6—OH
22.	—	A	Me	C_6—OMe
23.	Harmol	A	Me	C_7—OH
24.	Harmine Banisterine Yajeine Telepathine Leucoharmine	A	Me	C_7—OMe
25.	Ruine	A	Me	C_7—OMe; C_8—OGlc
26.	Harmalan	B	Me	—
27.	—	B	Me	C_6—OMe
28.	Harmalol	B	Me	C_7—OH
29.	Harmaline Harmidine	B	Me	C_7—OMe
30.	Dihydroruine	B	Me	C_7—OMe; C_8—OGlc

Table 1.-*Continued*



A



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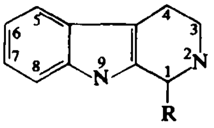
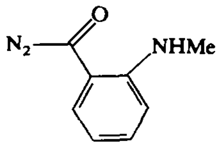


C

No.	Trivial name	Ring system	R	Additional substituents and their location
31.	Tetrahydroharman Eleagnine	C	Me	—
32.	—	C	Me	N_2—OH
33.	Leptocladine	C	Me	N_2—Me
34.	—	C	Me	N_2—Me; C_6—OMe
35.	—	C	Me	N_2—Me; C_7—OH
36.	—	C	Me	N_2—COMe; C_7—OCOMe
37.	—	C	Me	C_3—COOH
38.	—	C	Me	C_5—OMe
39.	Shepherdine	C	Me	C_6—OH
40.	—	C	Me	C_6—OMe
41.	Tetrahydroharmol	C	Me	C_7—OH
42.	Tetrahydroharmine Leptaflorine	C	Me	C_7—OMe
43.	—	A	COMe	—
44.	—	A	COMe	C_3—COOMe
45.	—	A	COMe	C_4—OMe
46.	Acetyl norharmine	A	COMe	C_7—OMe
47.	—	A	$CONH_2$	—
48.	Harmic amide	A	$CONH_2$	C_7—OMe
49.	—	A	COOMe	—
50.	Harmic acid ester	A	COOMe	C_7—OMe
51.	—	A	CH_2OH	—
52.	Harmalinic acid	B	COOH	C_7—OMe
53.	Pavettin	A	$CH{=}CH_2$	—
54.	Dehydrocrenatin	A	$CH{=}CH_2$	C_4—OMe
55.	—	A	$CH{=}CH_2$	C_4—OMe; C_7—OMe
56.	Dehydrocrenatidine	A	$CH{=}CH_2$	C_4—OMe; C_8—OMe
57.	—	A	$CH{=}CH_2$	N_9—OMe
58.	Crenatine	A	CH_2—Me	C_4—OMe
59.	Crenatidine	A	CH_2—Me	C_4—OMe; C_8—OMe
60.	Pyridindolol	A	$CH(OH)CH_2OH$	C_3—CH_2OH
61.	—	C	$CH_2CH(Me)_2$	—
62.	Borrerine	C	$CH{=}C(Me)_2$	—
63.	Rhetsinin	C	=O	
64.	—	C	=O	C_7—OMe

plants. It is of interest to note also that, whilst C_7-alkyl substitution is common in the naturally occurring simple β-carbolines, the correspondingly substituted indolalkylamines (C_6 of tryptamine) have only been reported in two plant species [128, 230]. This raises questions concerning the metabolism of both indolalkylamines and simple β-carbolines which would merit further investigation.

The similarity in chromatographic and spectral characteristics of simple β-carboline homologues and the ease of formation and structural modification which may take place in crude plant extracts [98, 185, 191] necessitates the use of caution in their identification in plant tissues. It is possible that reports of some of the naturally occurring forms found in plants (Table 2) represent artefacts of this kind.

In addition to their natural occurrence in plants, simple β-carbolines have been found in a number of other sources. Their presence in tobacco smoke [27–32], marijuana smoke [33], charred insects [34] and well-cooked foodstuffs [35, 36] reflects their formation from tryptophan by pyrolysis [27]. Their presence has

Table 2. Distribution of simple β-carboline alkaloids in fungi and angiosperms

Plant	Compound*	Reference
FUNGI		
Amanita muscaria (L. ex Fr.) Hooker	37	[61]
Coriolus maximus	11	[238]
Streptomyces alboverticillatus	60	[62]
ANGIOSPERMS		
Apocynaceae		
Amsonia tabernaemontana Walt.	24, 28, 31	[63, 64]
Apocynum cannabinum L.	28	[63]
Aspidosperma exalatum Monachino	15	[66]
A. polyneuron Müll. Arg.	15	[65]
Ochrosia nakaiana Koidz	11	[68]
Pleiocarpa mutica Benth.	49	[67]
Bignoniaceae		
Newbouldia laevis Benth. & Hook. f.	11	[69]
Calycanthaceae		
Calycanthus occidentalis Hook. & Arn.	24	[64, 70]
Chenopodiaceae		
Hammada leptoclada (M. Pop) M. Iljin	6, 31, 33	[71–75]
Kochia scoparia (L.) Schrad.	11, 24	[76]
Combretaceae		
Guiera senegalensis Lam.	11, 31	[77]
Cyperaceae		
Carex brevicollis DC.	11, 17, 18, 19, 20 23, 24	[78–88]
C. parva Nees.	17	[87]
Elaeagnaceae		
Elaeagnus angustifolia L.	6, 11, 23, 26, 31, 35, 41	[64, 70, 89–96]
E. commutata Bernh.	61	[97]
E. hortensis M. B.	31	[89]
E. orientalis L.	31	[89]
E. spinosa L.	31	[89]
Hippophae rhamnoides L.	11, 23, 28	[63, 96]
Shepherdia argentea Nutt.	41	[98]
S. canadensis Nutt.	39, 41	[98]
Gramineae		
Arundo donax L.	31, 33	[99, 116]
Festuca arundinacea Schreb.	1, 11	[106]
Lolium perenne L.	1, 11	[106]
Phalaris aquatica L.	6, 7	[105]
P. arundinacea L.	6, 7, 8	[100–104]
Lauraceae		
Nectandra megapotamica (Sprg.) Chodat & Hassler	7	[107]
Leguminosae		
Acacia baileyana F. Muell.	31	[110]
A. complanata A. Cunn.	31, 33	[109]
A. simplicifolia Schinz & Guillaumin	6	[108]
Anadenanthera peregrina (L.) Speg	6, 7, 34	[113–115]
Burkea africana Hook.	11, 26, 31	[111, 112]
Desmodium gangeticum DC.	2, 33	[119, 120]
D. gyrans DC.	33	[116]
D. pulchellum Benth. ex Baker	2, 7, 10, 11, 33, 40,	[116–118]
Mucuna pruriens DC.	22	[116]
Petalostylis labicheoides R. Br.	31	[121, 122]

Table 2.—*Continued*

Plant	Compound*	Reference
Prosopis nigra (Gris.) Hieron.	11, 31	[123]
Loganiaceae		
Strychnos melinoniana Baill.	13	[124]
S. usambarensis Gilg.	11	[125]
Malpighiaceae		
Banisteriopsis argentea Spring ex Juss.	24, 29, 33, 38, 42	[116, 147, 148]
B. caapi (Spruce ex Griseb.) Morton	14, 23, 24, 29, 42, 46, 48, 50, 52, 64	[126–141]
B. inebrians Morton	23, 24, 29, 42	[142–145]
B. lutea Ruiz	24	[155]
B. rusbyana (Niedenz) Morton	6	[149]
Banisteriopsis spp.	23, 24, 29, 42	[128, 146]
Cabi paraensis Ducke	24	[150, 151]
Myristicaceae		
Gymnacranthera paniculata (A. DC) Warb.	6	[154]
Virola cuspidata (Benth) Warb.	22, 27, 40	[152, 153]
V. rufula (A. DC) Warb.	7	[114, 115]
V. theidora Warb.	6, 7	[114, 115]
Ochnaceae		
Testulea gabonensis Pellegr.	6	[156]
Palmae		
Plectocomiopsis geminiflorus Becc.	9	[157]
Papaveraceae		
Meconopsis horridula Hook. f. & Thoms.	7	[158]
M. napaulensis DC.	7	[159]
M. robusta Hook. f. & Thoms.	7	[158]
M. rudis Prain	7	[158, 159]
M. paniculata (D. Don) Prain	7	[158]
Papaver rhoeas L.	7	[237]
Passifloraceae		
Passiflora actinea Hook.	11	[172]
P. alata Ait.	11	[172]
P. alba Link et Otto	11	[166]
P. bryonioides H. B. K.	11	[166, 170]
P. capsularis L.	11	[166]
P. caerulea L.	11	[171]
P. decaisneana Nichol	11	[171]
P. edulis Sims	11	[166, 171]
P. eichleriana Mast.	11	[172]
P. foetida L.	11	[171]
P. incarnata L.	11, 23, 24, 28, 29	[64, 160–171]
P. quadrangularis L.	11	[166]
P. ruberosa?	11	[166]
P. subpeltata Orteg.	11	[171]
P. warmingii Mast.	11	[171]
Polygonaceae		
Calligonum minimum Lipski	11, 12, 31, 32	[173–175]
Rubiaceae		
Borreria verticillata G. F. W. Mey	62	[187]
Leptactinia densiflora Hook. f.	31, 42	[180, 181]
Nauclea diderrichii	11, 16, 49	[185, 186]
Ophiorrhiza japonica Blume	11	[182]
Pauridiantha callicarpoides Bremek	11	[184, 189]

Table 2.—*Continued*

Plant	Compound*	Reference
P. dewevrei Bremek	11	[184]
P. lyalli	11	[190]
P. viridiflora Hepper	11	[184]
Pavetta lanceolata Eckl.	53	[183]
Psychotria carthaginensis Jacq.	6	[128]
P. viridis R. et P.	6	[128]
Simira klugii Standl.	11	[176–179]
S. rubra K. Schum.	11	[176–179]
Uncaria attenuata Korth.	11	[188]
U. canescens Korth.	11	[188]
U. orientalis Guill.	11	[188]
Rutaceae		
Araliopsis tabouensis Aubrév. & Pellegr.	63	[195]
Flindersia laevicarpa C. T. White & Francis	26	[191]
Xanthoxylum rhetsa DC.	63	[192–194]
Sapotaceae		
Chrysophyllum lacourtianum De Wild.	1, 51	[196]
Simaroubaceae		
Ailanthus malabarica DC.	43, 45, 47, 49, 54 56, 58, 59	[202]
Perriera madagascariensis Courchet	55	[197]
Picrasma ailanthoides Planchon	49	[201]
P. crenata (Vell) Engl.	49, 58, 59	[198, 199]
P. excelsa (Schwartz) Planchon	57	[203]
P. javanica Blume	54	[200]
Solanaceae		
Vestia lycioides Willd.	44	[204]
Symplocaceae		
Symplocos racemosa Roxb.	11	[205, 206]
Tiliaceae		
Grewia mollis Juss.	21	[207]
Zygophyllaceae		
Fagonia cretica L.	11	[208]
Nitraria schoberi L.	?	[209]
Peganum harmala L.	23, 24, 25, 28, 29, 30, 42	[96, 210–234]
Tribulus terrestris L.	11, 23, 24	[64, 96, 161]
Zygophyllum fabago L.	11, 23, 24	[64, 161, 235, 236]

* See Table 1 for structure.

also been reported in detached tobacco leaves [27], plant cell cultures containing high levels of tryptophan [40], saké [37, 38] and wine [39]. The presence of simple 1,2,3,4-tetrahydro-β-carbolines has been recorded in mammalian tissues [41–43] and their formation by the condensation of tryptamine with aldehydes formed as by-products of *in vitro* metabolism of animal tissue preparations has also been demonstrated [44]. The simple β-carbolines in passion-fruit juice [45, 46] and snuffs and decoctions prepared by South American Indian tribes [115, 128, 234] derive from their endogenous occurrence in botanical sources used for their preparation.

The sedative properties ascribed to passion-fruit juice [47] and the hallucinogenic properties of snuffs and decoctions prepared from species containing simple β-carbolines reflect their pharmacological activity. The simple β-carbolines have been found to act principally on the central nervous system and muscle [48]. Biochemical properties exhibited by these alkaloids in animal tissues include monoamine oxidase inhibition, monoamine uptake blockade and ion transport inhibition [49–51]. The simple β-carbolines have been used therapeutically, in the treatment of post-encephalitic Parkinsonism, a practice now discontinued [52], and in ecbolic applications [48].

Recent interest in these alkaloids has been stimulated by reports of their ability to potentiate mutagenicity of both known mutagens and non-mutagens in *Salmonella* cultures [53]. This has been shown to be, in part, due to their influence on the metabolism of the mutagens [54, 55]. However, the operation of a more direct mutagenic action [56] is supported by studies providing evidence of the intercalation of these bases into nucleic acids [57–59].

The simple β-carbolines have a widespread taxonomic distribution (Table 2) and this undoubtedly reflects their relative ease of formation from precursors which also have a widespread distribution. Their frequent occurrence as minor compounds in species containing more complex indole alkaloids justifies a view of their presence in these species as non-enzymatic by-products of tryptophan metabolism. However, in certain genera they represent the major group of alkaloids present and in levels which suggest enzyme-catalysed formation. Whilst the occurrence *per se* in a species is of limited taxonomic value, the presence of certain simple β-carbolines may constitute a useful characteristic. The presence of a C_4-substituted series in the Cyperaceae, C_6 in the Myristicaceae and C_7 in the Malpighiaceae and Zygophyllaceae might be provisionally considered in this way.

The selective advantages bestowed upon plant species by the presence of alkaloids is a field which provides ample speculation, but in addition to adverse pharmacological effects on predators, a role for the simple β-carbolines in defense against virus infection and as protozoacides [60] might be considered. It is not clear if the biochemical properties manifested in animal tissues, especially in relation to monoamine metabolism, are functional in plants.

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