Chemistry in Eriocaulaceae

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Eriocaulaceae is a pantropical family that comprises about 1100 species distributed in 11 genera. The infrafamilial relationships are still unsatisfactorily resolved, because of the tiny flowers and generalized morphology, which makes the taxonomy very difficult. Flavonoid and naphthopyranone profiles have proved to be important in order to contribute to the alignment of genera into the family. We here present a survey of the chemical data of Eriocaulaceae with a discussion about their contribution to the taxonomy of Eriocaulaceae.

Key words: Eriocaulaceae, Flavonoids, Chemotaxonomy

Introduction

Eriocaulaceae is a pantropical, predominantly herbaceous monocotyledonous family, comprising around 1100 species in 11 genera (Giulietti *et al.*, 2000; Sano, 2004). It is a common and diagnostic component of the herbaceous rocky outcrops vegetation of Brazil called "campos rupestres", that occurs above 900 m height. Plants of Eriocaulaceae live in sandy areas from swamps to drier soils and most of the species occurs mainly in Brazil (Giulietti and Hensold, 1991). Molecular and morphological studies characterized this family as a monophyletic group (Giulietti *et al.*, 1995, 2000; Bremer, 2002) that is recognized by the following synapomorphies: very small, unisexual, white flowers, in dense capitulae, with only one ovule per locule, and spiraperturate pollen.

In spite of the monophyly of the group, infrafamilial relationships are still unsatisfactorily resolved (Giulietti and Hensold, 1991; Giulietti *et al.*, 1995): *Paepalanthus* Mart., the largest genus, is polyphyletic (Giulietti *et al.*, 2000).

Chemical investigation of some species has been shown to be useful to taxonomically characterize the delimitation of several taxa. The first report about chemistry of Eriocaulaceae was published on the species of *Eriocaulon* L. by Bate-Smith and Harborne (1969), who found the flavonols quercetagetin, gossypetin and patuletin in the methanolic extract of the leaves. Dokkedal and Salatino (1992) found several flavones (luteolin *C*-glycoside, luteolin 7-*O*-triglucoside, luteolin 7-*O*-diarabinoside, nepetin, nepetin 7-*O*-glucoside and nepetin 7-*O*-arabinoside) in the methanolic extract of the leaves of six *Leiothrix* species. From those data, the presence of different classes of flavonoids clearly distinguished *Leiothrix* Ruhland (= flavones) from *Eriocaulon* (= flavonols). Mayworm and Salatino (1993) investigated four species of *Paepalanthus* and found mainly 6-oxygenated flavonols (beyond flavones) in *P. hilairei* Koern. [= *Actinocephalus bongardii* (A – St. Hil.) Sano]. Flavone glycosides were found in 22 taxa of *Syngonanthus* (Ricci *et al.*, 1996). This data suggested a close alignment between *Leiothrix*/*Syngonanthus* and distinguished this group from *Eriocaulon*/*Paepalanthus*. Besides flavonoids, Salatino *et al.* (1990) reported the contents of soluble phenolic compounds from the capitulae of eight species of Eriocaulaceae. Ho and Chen (2002) identified new flavan and hispidulin derivatives from the capitula of *Eriocaulon buergerianum* Koern.

Vilegas *et al*. (1990) investigated the chloroform extract of the capitula from *Paepalanthus bromelioides* Silveira and *P. vellozioides* Koern. and found a new isocumarin, named paepalantine. Paepalantine proved to be potent as antibiotic, cytotoxic and mutagenic (Varanda *et al.*, 1997). Since that, our group has been extensively investigating the chemistry of Eriocaulaceae, which is in course up to now. Besides flavonol and naphthopyranone derivatives, Andrade *et al*. (1999) described two new acyl glycosylated flavonoids in five species of nowadays *Actinocephalus* (cited as *Paepalanthus* sect. *Actinocephalus*). The presence of these compounds showed the qualitative similarity between the species belonging to this taxon. These data were a very important contribution when Sano (2004) recognized *Actinocephalus* as a new genus of Eriocaulaceae. Vilegas *et al*. (1999a) reported the presence of quercetagetin 7-methyl ether glycosides from the leaves of *Paepalanthus* subg. *Platycaulon* Ruhland. In another work, Vilegas *et al*. (1999b) described new naphthopyranone glycosides from species of *P.* subg. *Platycaulon*. Coelho *et al*. (1999a, b) found luteolin *O*- and *C*-glycosides in the capitula of *Syngonanthus bisulcatus* Ruhland. Besides eight known apigenin and luteolin *O*- and *C*-glycosides, Santos *et al*. (2001) isolated several xanthones from *Leiothrix* species. Andrade *et al*. (2002) reported the presence of 6-methoxy flavonols in the capitula of *Paepalanthus macropodus* Ruhland, which also produced naphthopyranones and flavonol derivatives (Vilegas *et al*., 1999a). Santos *et al*. (2002) isolated four known flavonoids and the new 6-methoxyquercetin-3-*O*-(6″-*E*-feruloyl)-*β*-D-glucopyranoside from the aerial parts of *P. polyanthus* Bong. [= *Actinocephalus polyanthus* (Bong.) Sano].

In face of the great contribution of the chemical information on the taxonomy of Eriocaulaceae, the aim of this work is to increase the knowledge of the chemistry of the Eriocaulaceae in total as well as to support its classification.

Results and Discussion

We here present the substances isolated from leaves, scapes and/or capitulae of Eriocaulaceae species. Tables I–III present the distribution of substances found in Eriocaulaceae species. Isolated compounds are presented in Figs. 1–3.

Table I. Distribution of flavones in Eriocaulaceae.

Taxon	Flavone															
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**	**16**
Paepalanthus																
P. subg. Xeractis																
P. chlorocephalus	xxx						xxx									
P. argenteus var. argenteus		xxx	xxx													
P. sect. Actinocephalus																
P. polyanthus																
P. hilairei																
P. robustus					x											
P. ramosus																
P. denudatus																
P. microphyllus																
P. brachypus																
P. subsect. *Aphorocaulon*																
P. macrocephalus																
P. sect. *Diphyomene*																
P. speciosus																
P. subg. *Platycaulon*																
P. vellozioides																
P. latipes																
P. bromelioides		x														
P. macropodus																
P. planifolius					xx	xx			xx			xx				
Syngonanthus																
S. bissulcatus				xx		xxx	xxx	xxx								
Leiothrix																
L. curvifolia		xx		xx												
L. flavescens						xx	xx							xx		xx

Refer to Fig. 1 for the structures of the compounds. x, Trace amount; xx, intermediary content; xxx, majority.

Table II. Distribution of flavonols, isoflavones, phenolic compounds and naphthopyranones in Eriocaulaceae.

Taxon	Flavonol																	
	17	**18**	**19**	**20**	**21**	**22**	**23**	**24**	**25**	**26**	**27**	**28**	**29**	**30**	**31**	**32**	**33**	**34**
Paepalanthus																		
P. subg. *Xeractis*																		
P. chlorocephalus	xx		xx															
P. argenteus var. *argenteus*								xx								xx		
P. sect. *Actinocephalus*																		
P. polyanthus	xxx			xx	xx		xxx	xx	x	xx								
P. hilairei	xxx	xxx	x		xx				xx	xx								
P. robustus		xxx	xx	xx			xx		x									
P. ramosus	xxx	xxx	x	xx	xx													
P. denudatus	xx	xx	xxx				x											
P. microphyllus																		
P. brachypu	xx								xxx									
P. subsect. *Aphorocaulon*																		
P. macrocephalus	x		xx					x										x
P. sect. *Diphyomene*																		
P. speciosus																		
P. subg. *Platycaulon*																		
P. vellozioides												xx	xx	xx	xx	xx	xx	
P. latipes												xx	xx	xx	xx	xx	xx	
P. bromelioides								xx				xx		xx				xx
P. macropodus	xx					xx		xx			xx							
P. planifolius		xx											xx					
Syngonanthus																		
S. bissulcatus																		
Leiothrix																		
L. curvifolia																		
L. flavescens																		

Taxon	Naphthopyranone																
	35	**36**	**37**	**38**	**39**	**40**	**41**	**42**	**43**	**44**	**45**	**46**	**47**	**48**	**49**	**50**	**51**
Paepalanthus																	
P. subg. *Xeractis*						x		x					x		x		
P. chlorocephalus																	
P. argenteus var. *argenteus*																	
P. sect. *Actinocephalus*																	
P. polyanthus																	
P. hilairei					xx	xx											
P. robustus					xx	xx											
P. ramosus			xx		xx	xx											
P. denudatus						xx	xx										
P. microphyllus				xxx	xxx	xxx				xxx	xxx	xxx					
P. brachypu																	
P. subsect. *Aphorocaulon*																	
P. macrocephalus					xx			x	x				x	xx			
P. sect. *Diphyomene*																	
P. speciosus				xxx	xx		xx						xx				
P. subg. *Platycaulon*																	
P. vellozioides				xxx		xxx							xx	xx	xx		
P. latipes				xxx		xxx							xx	xx	xx		
P. bromelioides				xxx	xxx	xxx										xxx	
P. macropodus																	
P. planifolius				xxx	xxx	xxx									xx	xx	xxx
Syngonanthus																	
S. bissulcatus																	
Leiothrix																	
L. curvifolia		xx															
L. flavescens	xx																

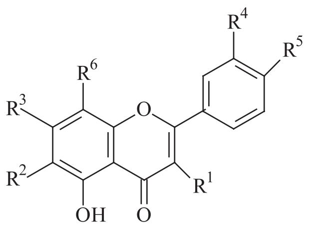
Refer to Figs. 1 and 2 for the structures of the compounds. x, Trace amount; xx, intermediary content; xxx, majority.

Table III. Distribution of xanthones in Eriocaulaceae.

Taxon	Xanthone		
	52	**53**	**54**
Leiothrix			
L. curvifolia		xxx	xxx
L. flavescens	xxx		

Refer to Fig. 3 for the structures of the compounds. x, Trace amount; xx, intermediary content; xxx, majority.

Table I demonstrates that flavone derivatives are frequent in *Paepalanthus* subg. *Xeractis* (Koern.) N. Hensold (*P. argenteus* var. *argenteus* Koern., *P. chlorocephalus* Silveira), as well as *Syngonanthus* and *Leiothrix* species. 6-Oxygenated flavonols are characteristic of *Actinocephalus* [*A. polyanthus, A. robustus* Silveira*, A. denudatus* (Koern.) Sano], while 7-methoxylated flavonols are more often found in *Paepalanthus* subg. *Platycaulon* Mart. (*P. vellozioides, P. latipes* Silveira, *P. bromelioides, P. macropodus*). Concerning naphthopyranones, Table II shows that these com-



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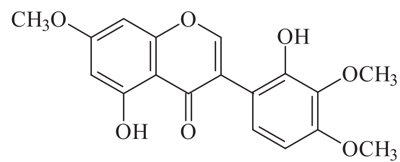

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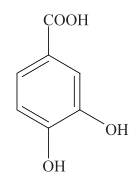


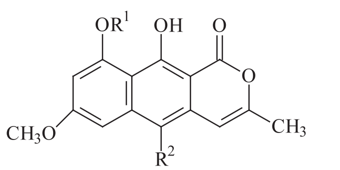
37

	R^1	R^2	R^3	R^4	R^5	R^6
1	H	OCH_3	OH	OH	OH	H
2	H	OCH_3	OH	OCH_3	OH	H
3	H	OCH_3	O-glu	OCH_3	OH	H
4	H	H	OH	OH	OH	H
5	H	OH	OH	OH	OH	H
6	H	OH	O-glu	OH	OH	H
7	H	glu	OH	OH	OH	H
8	H	glu	O-glu	OH	OH	H
9	H	H	OH	H	OH	H
10	H	OH	OCH_3	H	OH	H
11	H	OH	O-glu	H	OH	H
12	H	glu	OH	H	OH	glu
13	H	OH	O-glu	OH	O-glu	H
14	H	glu	OCH_3	OH	OH	H
15	H	OH	O-glu	OH	O-glu	H
16	H	OH	OH	OH	OH	OH
17	OH	OCH_3	OH	H	OH	H
18	O-glu	OCH_3	OH	H	OH	H
19	O-glu-*p*-coum	OCH_3	OH	H	OH	H
20	OH	OH	OH	OH	OH	H
21	OH	OH	O-glu	OH	OH	H
22	OH	OH	OH	OH	OH	OH
23	O-glu-*p*-coum	OCH_3	OH	OH	OH	H
24	OH	OCH_3	OH	OH	OH	H
25	O-glu	OCH_3	OH	OH	OH	H
26	O-rut	OCH_3	OH	OH	OH	H
27	O-glu-glu	OCH_3	OH	OH	OH	H
28	OH	OH	OCH_3	OH	OH	H
29	O-glu	OH	OCH_3	OH	OH	H
30	OH	OH	OCH_3	OH	O-glu	H
31	O-glu-glu	OH	OCH_3	OH	OH	H
32	O-glu-rham	OH	OCH_3	OH	OH	H
33	O-glu-glu-caff	OH	OCH_3	OH	OH	H
34	OH	OCH_3	O-glu	OCH_3	OH	H
35	O-glu-feru	OCH_3	OH	OH	OH	H

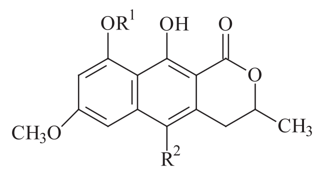
Fig. 1. Flavonoids isolated from Eriocaulaceae species.

pounds occur in large amounts in *P.* subg. *Platycaulon*, although they are also present in species belonging to *P.* sect. *Actinocephalus*, *P.* subsect. *Aphorocaulon* Ruhland and *P.* sect. *Diphyomene* Ruhland. Table III shows the occurrence of xanthone derivatives, which are exclusively from *Leiothrix* species.

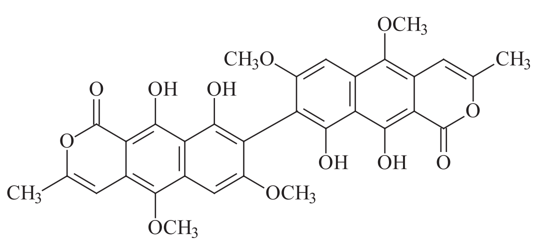
A cladistic analysis based on 49 predominantly morphologic characters (Giulietti *et al.*, 2000) suggested that *Paepalanthus* is polyphyletic and should be divided into smaller monophyletic genera. *Eriocaulon* is closely related to some groups of *Paepalanthus*, while *Leiothrix* and *Syngonanthus* emerge as sister groups. Based on biological, morphological, chemical and cytological characters, Sano (2004) proposed a new genus for Eriocaulaceae: *Actinocephalus* (Koern.) Sano, which in Ruhland's scheme (1903) was considered as a section in *Paepalanthus* subgenus *Paepalocephalus* and Körnicke (1863) considered as a subgenus of *Paepalanthus.* The other subgenera included in *Paepalanthus* (*P.* subg. *Platycaulon*, *P.* subg. *Thelxinoë* Mart., *P.* subg. *Psilandra* Mart.) display a considerably larger range of morphological variation at the species level, and therefore more detailed information about different aspects, such as morphology, anatomy, chemistry and cytology, is required. An exception is *Paepalanthus* subg. *Xeractis*, a group with 28 endemic species from the Serra do Espinhaço of Minas Gerais, Brazil (Hensold, 1988, 1998) and considered by Ruhland (1903) to be one of the few unquestionably natural infrageneric taxa. This group presents the flavone derivatives **1–3** (Fig. 1) as major constituents of *P. chlorocephalus* and *P. argenteus* var. *argenteus.* This result approaches *P.* subg. *Xeractis* to *Leiothrix* and *Syngonanthus.* These two genera are very close to each other and are chemically characterized by the presence of luteolin *O*- and *C*-glycosides as main constituents. On the other hand, the presence of the xanthones **52–54** (Fig. 3) and the absence of naphthopyranones in *Leiothrix* species allow us to distinguish between *Leiothrix* and *Paepalanthus*, since naphthopyranones (but not xanthones) are common in many *Paepalanthus* species (Table II). The occurrence of xanthones in *Leio-*



	R^1	R^2
38	H	OCH_3
39	glu	OCH_3
40	glu-allo	OCH_3
41	glu	H
42	glu-glu	H
43	H	H
44	glu-glu	OCH_3
45	ara-glu	OCH_3



	R^1	R^2
47	glu	H
48	glu-glu	H
49	allo-glu	H



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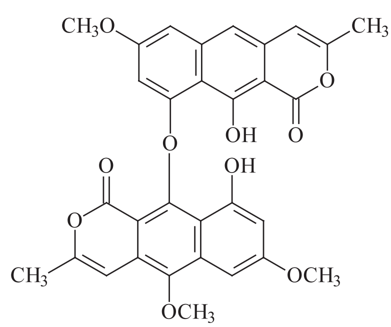
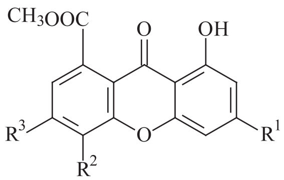


51

Fig. 2. Naphthopyranones isolated from Eriocaulaceae species.



	R^1	R^2	R^3
52	OH	OH	OH
53	OH	OH	OCH_3
54	OH	OCH_3	OCH_3

Fig. 3. Xanthones isolated from Eriocaulaceae species.

thrix contributes to distinguish this genus from *Syngonanthus*. Flavonoids also contribute to the chemical differentiation at the genus level, since *Paepalanthus* species produces 6- and/or 7-methoxyflavonol derivatives, whereas *Leiothrix* and *Syngonanthus* seem to produce mainly flavone derivatives (Santos *et al.*, 2001, Dokkedal and Salatino, 1992; Ricci *et al.*, 1996).

The chemical profile of *P. macrocephalus* Koern. (which belongs to *P.* subsect. *Aphorocaulon*) presents naphthopyranones (paepalantine derivatives and dihydronaphthopyranones **39**, **42**, **43**, **47**, **48**, Fig. 2), in addition to other compounds, such as the acylated flavonols and flavonol glycosides **17**, **19**, **24**, **34** (Fig. 1). The presence of naphthopyranones in *P.* subsect. *Aphorocaulon* is in agreement with the cladistic analysis (Giulietti *et al.*, 2000), where this taxon forms a sister group with *Actinocephalus* (which has a similar chemical profile).

Chemical data corroborate the phylogenetic hypothesis that *Paepalanthus* is polyphyletic and must be divided into a number of more natural groups. The chemical composition has proved to be an important taxonomic character, since the chemical profiles are quite distinct in the different taxa, although not necessarily exclusive. The occurrence of 6-methoxylated flavonoids as well as glycosylation at C-3 seems to be a major trend in *Paepalanthus*. The presence of naphthopyranones, mainly paepalantine derivatives, is characteristic of *P.* subg. *Platycaulon,* together with minor amounts of dihydronaphthopyranones and flavonol derivatives. The proximity of *P.* subg. *Platycaulon* to *P.* subsect. *Aphorocaulon, P.* serie *Dimeri* Ruhland*, P.* sect. *Diphyomene* Ruhland and *Actinocephalus* is also supported by the similarity between their chemical profiles, which include flavonol and naphthopyranone derivatives in all of them (see Table II). This proximity indicates probably a common evolutionary history, as shown by the cladistic analysis of Giulietti *et al.* (2000). *Actinocephalus*, on the other hand, can be distinguished from the other taxa by the presence of acylated flavonol glycosides as well as glycosylated derivatives of paepalantine. These chemical characters reinforce *Actinocephalus* as a distinct genus separated from *Paepalanthus* as proposed by Sano (2004). In *P.* subsect. *Aphorocaulon*, major constituents are monoglycosylated paepalantine, desmethoxypaepalantine and dihydronaphthopyranones, and in *P.* sect. *Diphyomene* these are mono- and diglycosylated paepalantine and glycosylated dihydronaphthopyranone. *Paepalanthus* subg. *Xeractis* is characterized by the presence of flavone derivatives and minor amounts of paepalantine, desmethoxypaepalantine and dihydronaphthopyranone derivatives. Therefore, the presence of these compounds in *P.* subg. *Xeractis* gives support for a close relationship between *P.* subg. *Xeractis* and *Leiothrix/Syngonanthus*, as demonstrated by the cladistic treatment (Giulietti *et al.*, 2000). Furthermore, this profile corroborates the proposition of Giulietti *et al.* (2000), who stated that *P.* subg. *Xeractis* is very distinct from *Paepalanthus*. The presence of flavones instead of flavonols in both *P.* subg. *Xeractis* and *Leiothrix/Syngonanthus* group is in agreement with the fact that flavones often appear to replace flavonols in the most advanced groups (Salatino *et al.*, 1990). Finally, xanthones can be useful chemical markers to distinguish between *Leiothrix* and *Syngonanthus* (Santos *et al.*, 2001).

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