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# Occurrence and Distribution of Polyphenolics in Species of *Deguelia* (Leguminosae)

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## Abstract

The species of *Deguelia* (Papilonoidae Leguminose) genus are recognized for producing polyphenolic secondary metabolities that are predominantly classified as isoflavonoids. The study about the distribution of these secundary metabolities in species of *Deguelia* genus is very important for understanding the chemosystematic at genus level. Additionally, it has been reported that polyphenolics isolated from *Deguelia* present many desirable biological effects against human diseases and agricultural pests. Thus, this paper reviewed the occurrence and distribution of polyphenolics in species of *Deguelia* and evidenced the biological properties of these compounds.

**Keywords:** *Deguelia* (Papilonoidae Leguminosae); Polyphenolics; Leguminosae; Papilionoideae

## Introduction

*Deguelia* genus belongs to Papilionoideae subfamily, Leguminosae family and comprises 20 species mainly distributed in South America [1,2]. Some of them are used by Amazon indigenous population due to its ictiotoxic properties, such as *Deguelia duckeana*, known as "cipó cururu" or "timbó" [3], *D. utilis* and *D. rufescens* var. urucu, commonly known as "timbó branco" and "timbó vermelho", respectively [4,5]. Their insecticide and ichthyotoxic activities are attributed to rotenoids presence in its roots [4]. *Deguelia* species are rich in polyphenolic which demonstrate many biological activities such as leishmanicidal [6], cytotoxic against *Artemia salina* [3], antitumoral [7], antioxidant [8], phytotoxicity [9], antimicrobial [10,11] and vasorelaxant effect [12], among others. In this context, the aim of this paper is to review *Deguelia* polyphenolics and biological potential of their species.

### *Deguelia* species distribution in Brazil

Seventeen brazilian *Deguelia* species are distributed into two sections, based on morphological characters, specially the number of eggs in the ovary: *Deguelia* sect. *Multiovulis* A.M.G. Azevedo comprises five species containing 7-15 eggs per ovary: *Deguelia costata* (Benth.) A. M. G. Azevedo & R. A. Camargo, *D. densiflora* (Benth.) A. M. G. Azevedo ex M. Souza, *D. hastchbachii* (Benth.) A.M.G. Azevedo, *D. longeracemosa* (Benth.) A.M.G. Azevedo and *D. spruceana* (Benth.) A. M. G. Azevedo & R. A. Camargo, while species with six eggs per ovary are allocated in *Deguelia* sect. *Deguelia*: *D. amazonica* Killip, *D. angulata* (Ducke) A.M.G. Azevedo & R.A. Camargo, *D. utilis* (A. C. Sm.) A. M. G. Azevedo, *D. scandens* Aubl., *D. negrensis* (Benth.) Taub. [Magalhães], *D. duckeana* A. M. G. Azevedo, *D. rariflora* (Mart. ex Benth.) G. P. Lewis & Acev.-Rodr., *D. urucu* (Killip & A. C. Sm.) A. M. G. Azevedo & R. A. Camargo, *D. nitidula* (Benth.) A. M. G. Azevedo & R. A. Camargo, *D. glaucifolia* A. M. G. Azevedo, *D. dasycalyx* (Harms) A. M. G. Azevedo & R. A. Camargo. Many of these species have *Lonchocarpus* sp and *Derris* sp. synonyms [1,2].

Among *Multiovulis* sect. species, three occur mainly in two brazilian regions: Southeastern, in the states Espírito Santo, Minas Gerais and Rio de Janeiro (*D. costata*, *D. hatchibachii* and *D. longeracemosa*) and Northern, in Amapá, Amazonas and Pará (*D. densiflora* and *D. spruceana*). However, *D. spruceana* was also found in the northern state of Maranhão [1,2].

The species *Deguelia* sect. is mainly distributed in the Northern states, being found in Pará, Roraima, Amapa, Rondônia, Amazonas and Acre. *D. nitidula* is one of the few *Deguelia* sect. species found in other regions such as Midwestern Goiás and Mato Grosso; Northeastern region in Maranhão and Piauí and Southeastern region in Minas Gerais and São Paulo. *D. scandens* is also found in Maranhão [1,2].

### *Deguelia* genus polyphenolics

*Deguelia* species are rich in polyphenolics secondary metabolites such as chalcones, stilbenes, isoflavonoids (rotenoids, isoflavones, 4-hydroxy-3-phenylcoumarins, isoflavan and isoflavanones) and flavonoids derivatives (flavanones and dihydroflavanols).

### Rotenoids

The review detected the presence of thirty four rotenoids in *Deguelia* species (Compounds **1-30** and **103-106**, Figure 1). *Deguelia* sect. species was mainly isolated in Brazilian and Peruvian Amazon forest: *D. utilis* [13,14], *D. negrensis* [15], *D. amazônica* [16] and Cubé resin, formed by mixing *D. utilis* and *D. rufescencs* var. urucu [17]. Thedehydrorotenone (1) was also the only rotenoid isolated from *Multiovulis* section *D. densiflora* species [7]. These compounds were predominantly found in the roots, followed by the stems and aerial parts of these species. Rotenone (**1**) was the most widely distributed in *Deguelia* species (Figure 1).

### Stilbenes

Currently, eighteen stilbenes (compounds **31-47** and **109**, Figure 1) and their derivatives have been isolated from *Deguelia* genus. The

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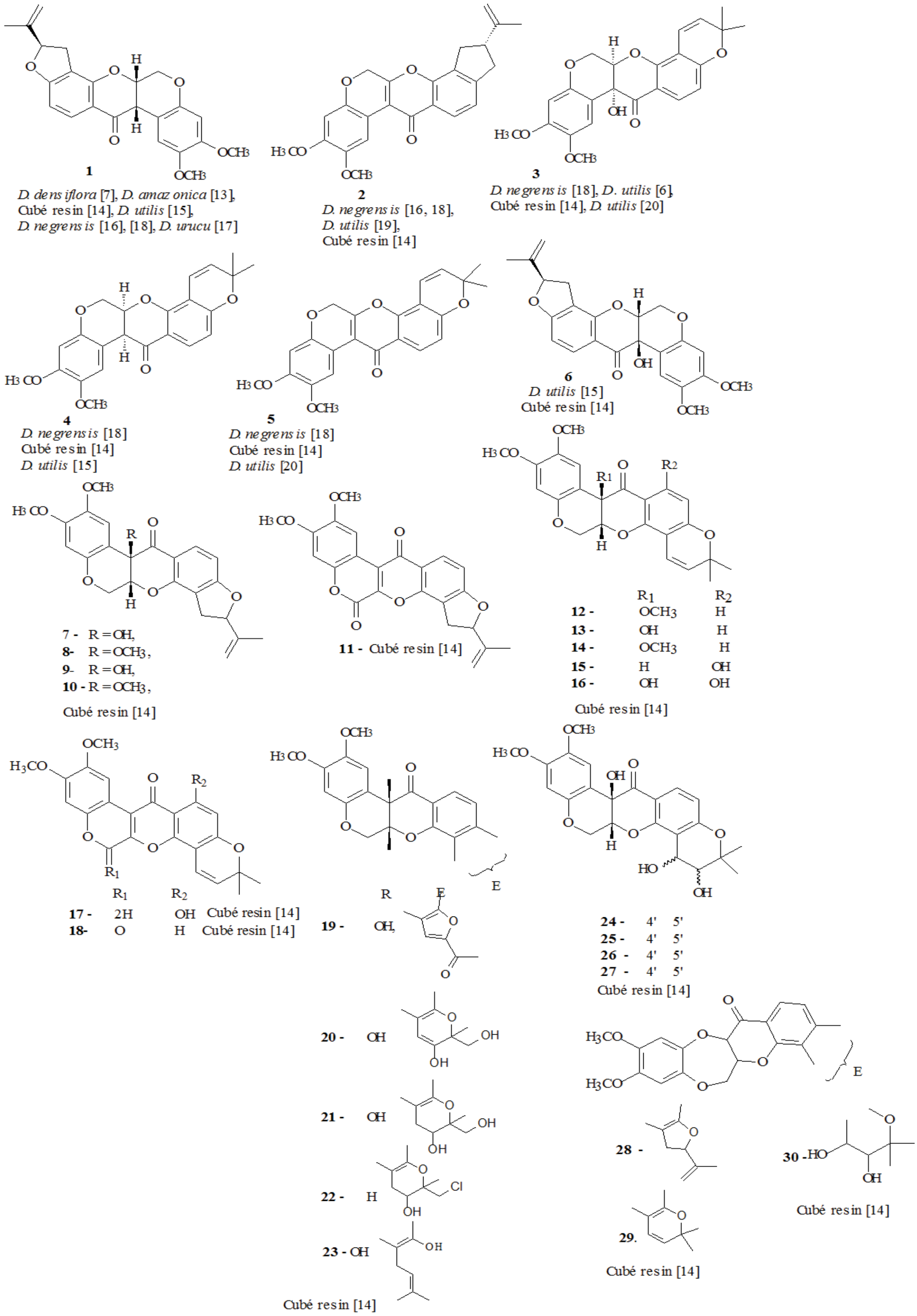
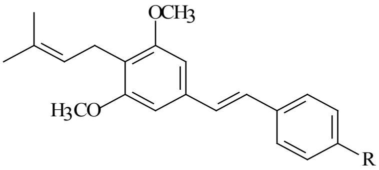
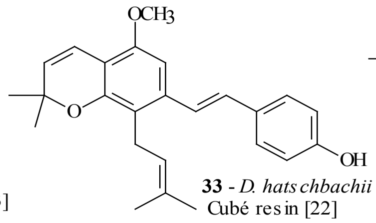
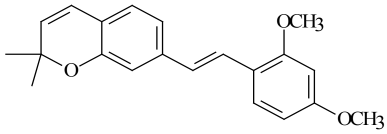

1
D. densiflora [7], D. amazonica [13], Cubé resin [14], D. utilis [15], D. negrensis [16], [18], D. urucu [17]
2
D. negrensis [16, 18], D. utilis [19], Cubé resin [14]
3
D. negrensis [18], D. utilis [6], Cubé resin [14], D. utilis [20]
4
D. negrensis [18]
Cubé resin [14]
D. utilis [15]
5
D. negrensis [18]
Cubé resin [14]
D. utilis [20]
6
D. utilis [15]
Cubé resin [14]
7 - R = OH,
8- R = OCH3,
9- R = OH,
10 - R = OCH3,
Cubé resin [14]
11 - Cubé resin [14]
R1 R2
12 - OCH3 H
13 - OH H
14 - OCH3 H
15 - H OH
16 - OH OH
Cubé resin [14]
R1 R2
17 - 2H OH Cubé resin [14]
18- O H Cubé resin [14]
R E
19 - OH,
20 - OH
21 - OH
22 - H
23 - OH
Cubé resin [14]
24 - 4' 5'
25 - 4' 5'
26 - 4' 5'
27 - 4' 5'
Cubé resin [14]
28 -
29.
Cubé resin [14]
30 -
Cubé resin [14]



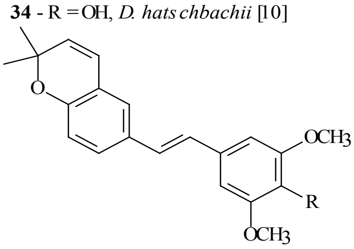

**31** - R = H, *D. nitidula* [17], *D. rariflora* [21]
**32** - R = $OCH_3$, *D. nitidula* [17], *D. duckeana* [3]
**34** - R = OH, *D. hatschbachii* [10]



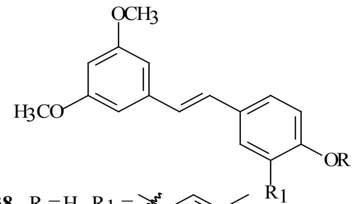

**33** - *D. hatschbachii* [10], Cubé resin [22]

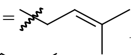
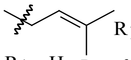


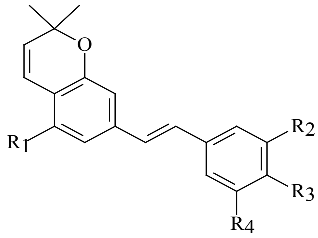

**35** - *D. spruceana* [25]



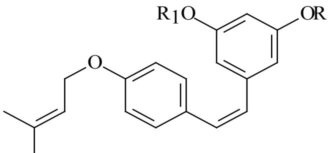

**36** - R = $OCH_3$, *D. rufescens* [9], *D. utilis* [4]
**37** - R = H, *D. rufescens* [5, 9], *D. utilis* [5, 20, 23]



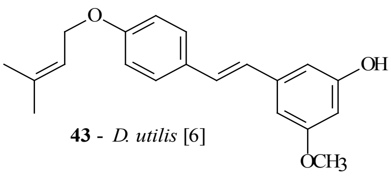

**38** - R = H, $R_1$ =  *D. utilis* [6], *D. rufescens* [9]
**39** - R =  $R_1$ = H *D. utilis* [6], *D. rufescens* [5, 9]
**40** - R = $R_1$ = H, *D. rufescens* [9]



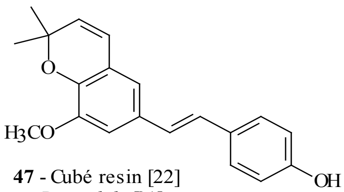

**44** - $R_1$ = $R_2$ = $OCH_3$, $R_3$ = OH, $R_4$ = H
**45**- $R_1$ = H, $R_2$ = $R_3$ = $R_4$ = $OCH_3$
**46** - $R_1$ = $OCH_3$, $R_2$ = $R_4$ = H, $R_3$ = OH
Cubé resin [22]



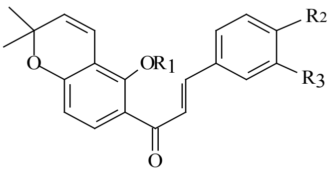

**41** - R = $CH_3$, $R_1$ = H
**42** - R = $R_1$ = $CH_3$
*D. utilis* [6]



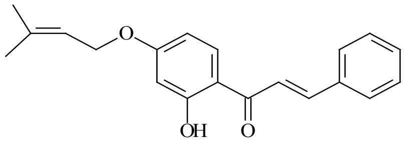

**43** - *D. utilis* [6]



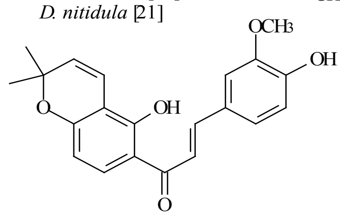

**47** - Cubé resin [22]
*D. nitidula* [21]



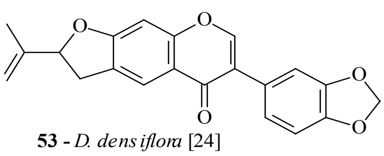

**48** - $R_1$ = $R_2$ = $R_3$ = H, *D. duckeana* [3]
**49** - $R_1$ = $R_3$ = H ; $R_2$ = OH, *D. duckeana* [3], *D. nitidula* [17], *Cubé resin* [22]
**50** - $R_1$ = H; $R_2$ = $R_3$ = OH, *D. nitidula* [17]



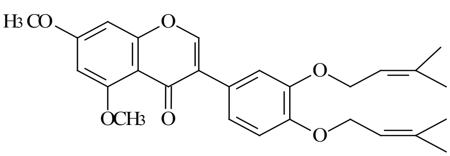

**51** - *D. duckeana* [3]



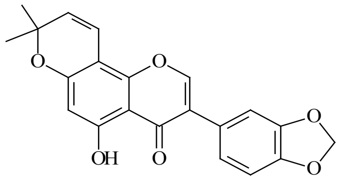

**52** - Cubé resin [22]



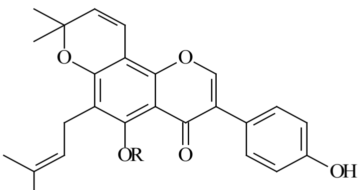

**53** - *D. densiflora* [24]



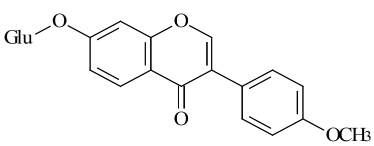

**54** - *D. densiflora* [24]



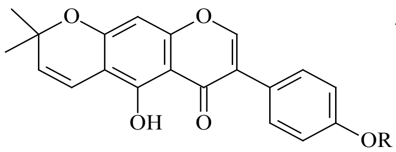

**55** - *D. spruceana* [25], *D. hatschbachii* [10], *D. longeracemosa* [11]



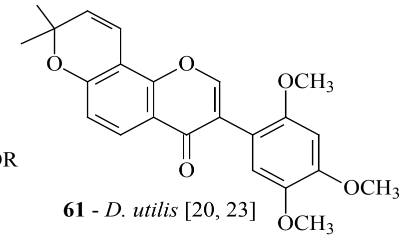

**56** - R = H, osajin, *D. longeracemosa* [11]
**57** - R = $CH_3$ , scandinone, *D. hatschbachii* [10]



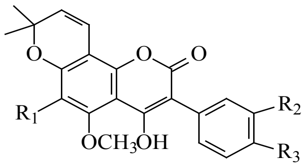

**58** - *D. longeracemosa* [11]

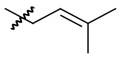


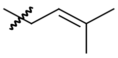
**59** - R= H, *D. densiflora* [7]
**60** - R= $CH_3$, *D. densiflora* [7]

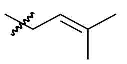


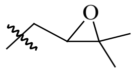
**61** - *D. utilis* [20, 23]

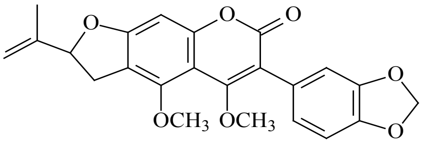


**65**- $R_1$=  ; $R_2$= H; $R_3$= OH,
*D. spruceana* [25], *D. hatschabachii* [10], *D. longeracemosa* [11]

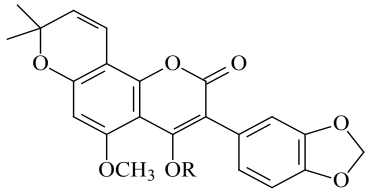
**66** - $R_1$=  ; $R_2$= OH; $R_3$= $OCH_3$
*D. longeracemosa* [11]

**67**- $R_1$=  ; $R_2$,$R_3$= –$OCH_2O$–
*D. longeracemosa* [11]

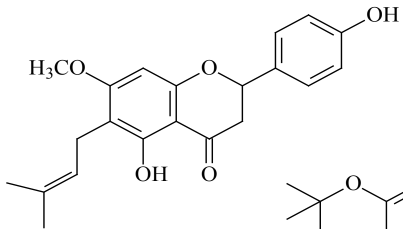
**68** - $R_1$=  ; $R_2$= H; $R_3$= OH
*D. longeracemosa* [11]



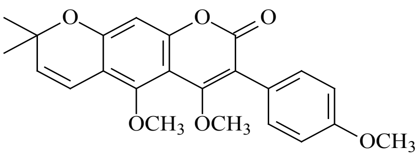
**62** - *D. densiflora* [24]



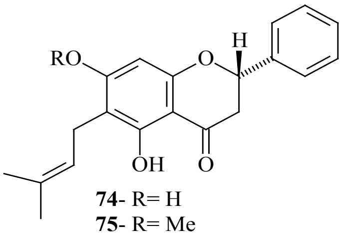
**63** - R= H, *D. spruceana* [25]
*D. longeracemosa* [11]
**64** - R= $CH_3$, *D. spruceana* [25]



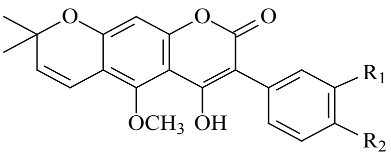
**80** - *D. hatschbachii* [10]

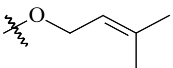


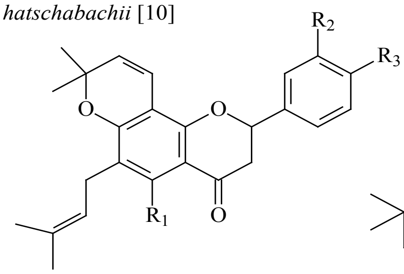
**73** - *D. hatschabachii* [10]



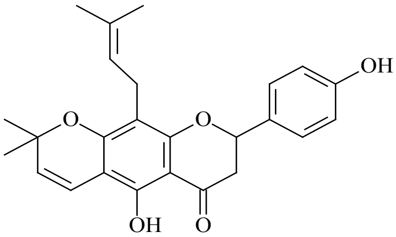
**74**- R= H
**75**- R= Me
*D. rariflora* [21]



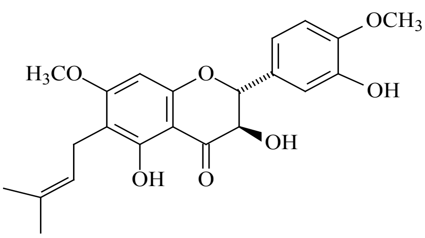
**69**- $R_1$= OH ; $R_2$= $OCH_3$, *D. longeracemosa* [11]
**70**- $R_1$= H ; $R_2$= 
*D. longeracemosa* [11]
**71**- $R_1$, $R_2$= –$OCH_2O$–. *longeracemosa* [11]
**72**- $R_1$= H ; $R_2$= $OCH_3$, *D. longeraccemosa* [11], *D. hatschabachii* [11]



**76**- $R_1$= OH; $R_2$= OMe; $R_3$= OMe,
**77**- $R_1$= OH; $R_2$= OH; $R_3$= OMe
**78**- $R_1$= OH; $R_2$= OMe; $R_3$= OH
**79**- $R_1$= H; $R_2$= OMe; $R_3$= OMe
Cubé resin [22]



**81** - *D. hatschbachii* [10]



**82**- *D. rufescens* [8, 12]

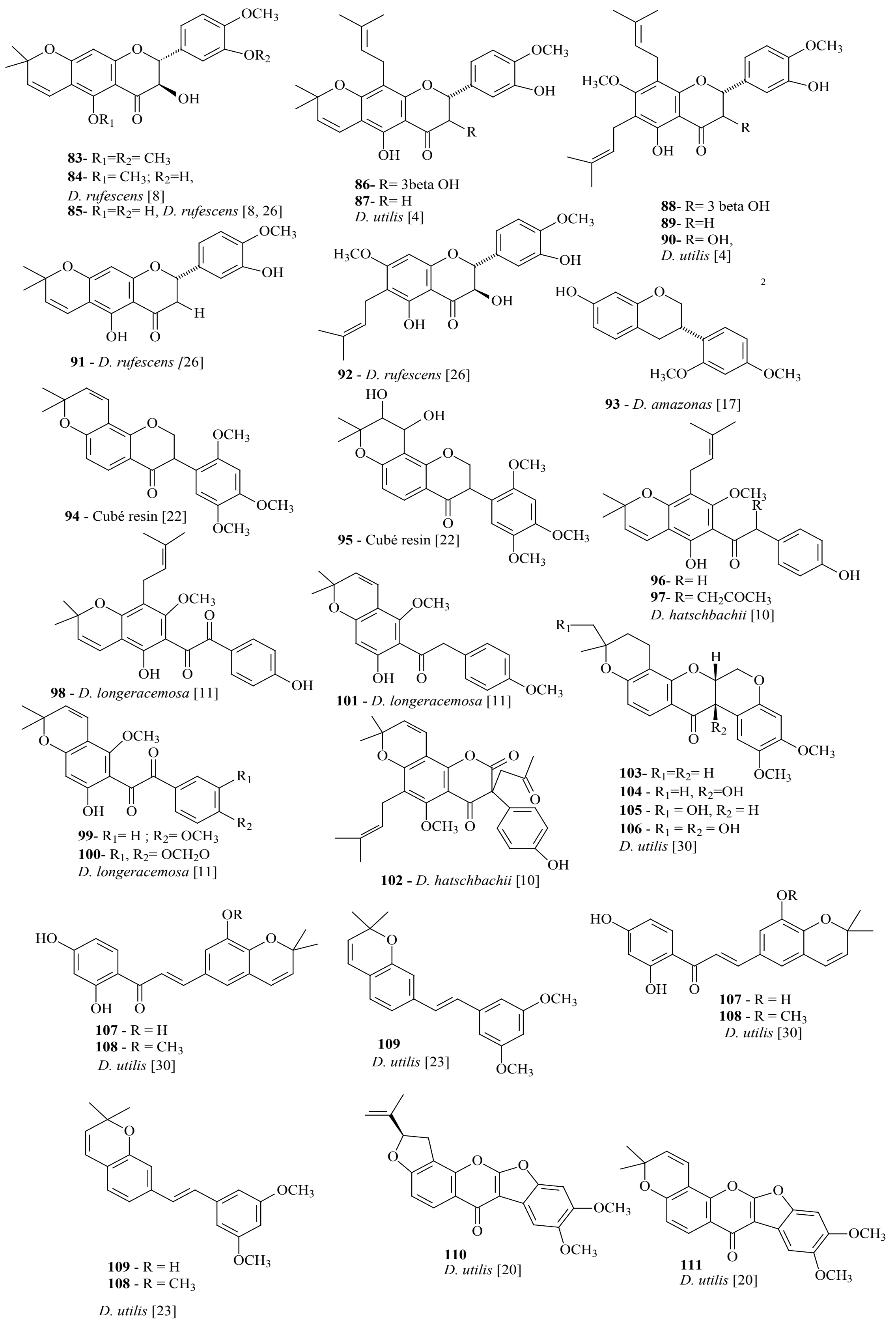



**Figure 1:** Molecular structures (**1-111**) of the polyphenolics found in Deguelia genus.

compounds (**31-33**) and (**35-47**) were isolated from *Deguelia* sect. species: *D. nitidula* [18], *D. rariflora* [19], *D. duckeana* [3], *D. utilis* [4,14], *D. rufescens* var. urucu [9] and Cubé resin (*D. utilis* and *D. rufescens* var. urucu) [20], while only three compounds were isolated from *Multiovulis* section species, (**33**) and (**34**), from *D. hatschbachii* [10] and compound (**35**) from *D. spruceana* [21]. These compounds were mainly found in roots and leaves of plants from Amazon forest. The exception was for *D. hatschbachii*, which was collected in Campinas, SP. The stilbenes (**31**), (**32**), (**33**), (**35**), (**36**), (**37**) and (**38**) were found in at least two different *Deguelia* genus species. Except for compound (**40**), all the other stilbene derivatives are prenylated.

## Chalcones

The occurrence of prenylated chalcones derivatives in ring B (compounds **48-52** and **107-108**, Figure 1) was only reported in *D. duckeana* [3], *D. nitidula* [18], Cubé resin (*D. utilis* and *D. rufescens* var. urucu) [20], all of them belonging to *Deguelia* sect. species.

## Isoflavones

A total of nine isoflavones (compounds **53-61**, Figure 1) have been reported in *Deguelia* plants. These ones were widespread in *D. densiflora* [22], *D. spruceana* [22], *D. hatschbachii* [10] and *D. longeracemosa* [11], all of *Multiovulis* sect. Only lonchocarpusone (**61**) was reported in *D. utilis* [23], *Deguelia* sect. The compounds predominant isolation was in the roots except for glabrescione (**53**) and glabrescione B (**54**) which was found in *D. densiflora* seeds.

## 4-Hydroxy-3-phenylcoumarins

The twelve prenylated derivatives in 4-hydroxy-3-phenylcoumarins ring A (compounds **62-73**, Figure 1) were only reported in *D. densiflora* [24], *D. spruceana* [25], *D. longeracemosa* [11] and *D. hatschbachii* [10], all of them belonging to the *Multiovulis* sect. These ones were mainly detected in the roots and showed lower occurrence in seeds as glabrescin (**62**) which was isolated from *D. densiflora* seeds [21].

## Flavanones and dihydroflavanols

Flavanones (**79-81**), (**87**) and (**89**) (Figure 1) and dihydroflavanols (**82-86**), (**88**) e (**90-92**) (Figure 1) were identified in *Deguelia* genus. Only these compounds have a flavonoid skeleton. The derivatives of flavanones (**75-80** and **82-92**) were reported in *D. rariflora* [19], *D. rufescens* var. urucu [8,12], *D. utilis* [4] and roots of Cubé resin (*D. utilis* e *D. rufescens* var. urucu) [20] from *Deguelia* sect. Yet, the compounds (**80**) and (**81**) were isolated from *D. hatschbachii Multiovulis* sect. roots [10]. Every compound structure showed preny and/or 2,2-dimethylchromene in carbon 6 and hydroxyl and/or methoxyl group in carbon 5, in ring A.

## Isoflavanones and isoflavan

These classes of secondary metabolites are rarely found in the *Deguelia* genus. The derivative of isoflavan (**93**) (Figure 1) was obtained from wood of *D. amazonas Deguelia* sect. [18]. Two derivatives of isoflavanones (**94**) and (**95**) (Figure 1) were reported in Cubé resin (*D. utilis* and *D. rufescens* var. urucu), both of *Deguelia* sect. [20].

## Ethanediones and deoxybenzoin

Oxidized derivatives of stilbene as ethanediones and deoxybenzoin were only reported only in *Multiovulis* sect. plants (Compounds **96-101**, Figure 1). The compounds (**96**) and (**97**) were isolated from *D. hatschbachii* [10] and (**98-101**) of *D. longeracemosa* roots [11].

## 4-hydroxy-3-phenylcoumarin derivative

The scandenin alkylated derivative in C-3 (**102**, Figure 1) was isolated from *D. hatschbachii* roots. This compound is $\beta$-keto tautomer which corresponds to C-3 substituent ester.

## Coumaronochromones

Only two derivatives of coumaronochromones (**110** and **111**, Figure 1) were isolated from roots of *D. utilis*, *Deguelia* sect. [14].

The most characteristic classes of secondary metabolites with predominant occurrence were distributed in two sections of *Deguelia* genus as rotenoid, stilbene and chalcone derivatives, in *Deguelia* sect. species and 4-hydroxy-3-phenylcoumarins, isoflavones, ethanediones and deoxybenzoin derivatives in *Multiovulis* sect. species. These are in agreement with the previous phytochemical studies on *Deguelia* species [10]. Rotenone (**1**) and scandenin (**65**) are the most representative compounds for the *Deguelia* and *Multiovulis* sections, respectively. The isoflavone derivatives are biosynthetic precursors of both rotenoids as 4-hydroxy-3-phenylcoumarins [24]. This demonstrates that species from both sections should be presumably endowed with strongly oxidizing enzymes [25]. Research about polyphenolic distribution in literature such as isoflavonoids and chalcones in species of a genus may help distinguish the evolution phase of its species [26].

## Biological potential of *Deguelia* species

*Deguelia* species, which come from Peruvian and Brazilian Amazon and also other parts of Brazil, had their biological potential evaluated on the basis of extracts and compounds (molecular structures of these compounds are in Figure 1) which have been isolated from different species of this genus. In this survey, ten experimental models used to evaluate the potential of the control *Deguelia* the application of biological activities were recorded. Roots ethanolic extract from *D. amazonica* presented 3.7 % of rotenone (**1**) (Figure 1) in its composition, showed toxicity to *Cerotoma arcuatus* (bean insect) when incorporated by contaminated leaves ingestion, by contact with contaminated surface and by topical application [16]. It is more effective for the first condition with adult mortality higher than 80% at 3.0% concentration v/v [13]. Flavonoids (**76-79**) and stilbenes (**44-46**) isolated from the roots of the natural insecticide Cubé resin [20] (*D. utilis* and *D. rufescncs var.* urucu*)*, showed inhibitory activity for NAD / ubiquinone oxidoreductase enzymes and ornithine decarboxylase (ODC) with $IC_{50}$ ranging from 4.9 to 0.68 and 7.4 to 3.9 μM, for flavonoids and stilbenes, respectively, which was the best result for flavonoid (**78**) to the two enzymes, followed by (**76**, **77** and **79**) (Figure 1). The presence of dimethylpyran ring seems to be a key factor for these compounds activity, just like the OH in C3 position seems to be the differentiating factor for (**78**). Besides these ones, Fang and Casida, in 1999 [17], isolated and evaluated other compounds in the same kind of Cubé resin, the rotenoids (**1**, **3**, **4** and **7**) and twenty-five derivatives (**2**, **10** to **30**) [14]. Rotenone (**1**) and deguelina (**4**) were the most active, showing $IC_{50}$ 0.0044 and 0.0069 μM, respectively. Still regarding enzymatic activity, Pereira et al. [5] reported lonchocarpene (**37**) and 3,5- dimethoxy -4' -O- prenyl -*trans*- stilbene (**39**) inhibitory action for in vitro α –glucosidase with $pIC_{50}$ of 5.68 ± 0,12 and 5.73 ± 00:08, respectively, target used to control type 2 diabetes. However, only (**39**) was able to reduce induced hyperglycemia in rats. Fang and Casida [20] also reported anti-tumor activity for flavonones (**49**, **77-79, 94** and **95**) and *in vitro* stilbenes (**44-46**) to MCF 7 and Hepa 1c1c7 cells strains. $IC_{50}$ ranged between 0.34 and 30.0 μM, in which compound (**94**) showed the best results 1.9 and 0.34 μM, for each of the strains, respectively. Current research aimed at fighting the tumor cell activities go through signaling mechanisms control. In this sense, Lui et al. [7] reported isoflavones inhibitory activity (**59**) and (**60**) regarding hypoxia inducing factor, HIF -1, for human breast tumor T47D cell, a key factor for solid tumors combat, showing $IC_{50}$ = 0.6 µM and 5.0 µM, respectively. For *Artemia salina* toxicity test Magalhães et al. [10] assessed the compounds (**65, 80** and **81**), finding $LC_{50}$ 4.37; 0.0004 and 13.1 µg $mL^{-1}$, respectively. Magalhães et al. [10,11] evaluated the antimicrobial activity in bioautography assay for compounds (**33**, **34**, **56**, **63**, **65**, **66**, **73**, **80**, **81**, **97**, **99**, **102**). Compounds (**65** and **66**) showed activity against *Staphylococcus aureus* and *Bacillus subtilis* and compounds (**63**, **71** and **98**) only for *B. subtilis*. Additionally, Lobo et al. [9] reported allelopathic compounds (**36-40**), which maximum inhibition germination did not exceed 20% of seeds from *Mimosa pudica*. However, when evaluated in pairs, the effect was higher, showing synergistic activity between compounds. On the other hand, Silva et al. [27-30] showed the opposite effect, lower activity for compounds (**83**, **84** and **85**) when evaluated in pairs. As antioxidant activity, Lobo et al. [8] assessed the compounds (**82-85**) by DPPH method and all compounds showed small activity. Compound (**82**) produced 40 ± 4.4 % of the relaxing effect on rat aorta at 100 µM concentration as shown by Mendes et al. [12]. Finally, it can still be reported the compounds neuroprotective activity (**86** to **90**) and (**36** and **37**) by Oliveira et al. [4].

## Conclusion

Based on these phytochemical studies results we suggest the presence of secondary metabolites as oxidized stilbenes derivatives such as ethanediones and deoxybenzoin only in *Multiovulis* species and chalcones derivatives only in *Deguelia* sect. indicating that species in *Multiovulis* is more evolved than in *Deguelia* sect. Moreover, it was evidenced that flavonoids, stilbenes, isoflavones and rotenoids, isolated from *Deguelia*, present high biological potential, thus highlighting the necessity of new researches in order to achieve a better understanding about the secondary metabolites of *Deguelia* genus.

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