



Review

Distribution of flavonoids in the Myristicaceae

Juan C. Martínez Valderrama *

Departamento de Química, Universidad Nacional de Colombia, A.A. 14490, Santafé de Bogotá, Colombia

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Dedicated to Professor Dr. Otto Richard Gottlieb for his 80th birthday

Abstract

During the past two decades the study of the Myristicaceae species has increased, and many flavonoids of different types have been isolated. This review discusses the flavonoids presently known in the Myristicaceae. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Myristicaceae; *Compsonura*; *Dialyanthera*; *Iryanthera*; *Knema*; *Myristica*; *Osteophloeum*; *Otoba*; *Pycnanthus*; *Viola*; Flavonoids

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1. Introduction

Interest in the Myristicaceae family was inspired by their ethnopharmacology, and a review describing the research on these plants (Gottlieb, 1979) clearly indicated the need for phytochemical and pharmacological studies of this family. The Myristicaceae family belongs to the Magnoliiflorae super-order, morphologically considered one of the most primitive of the Angiosperms, and comprises nineteen genera of trees and shrubs dis-

tributed in Asia (four genera), Africa and Madagascar (nine genera) and America (six genera). Fifty samples from among the six genera native to the American continent are housed in the Colombian National Herbarium, including five *Compsonura*, five *Dialyanthera*, eleven *Iryanthera*, two *Osteophloeum*, three *Otoba* and twenty-four *Viola*. We have collected material (fruit, leaves, bark and wood) from fifteen of these species in the Amazonian region, the Eastern Plains, the Middle Magdalena Antioqueño and the Chocó, and have isolated and identified flavonoids from eight of them. The presence of flavonoids in species of the Myristicaceae family is less common than in other families of Angiosperms, but the occurrence of certain classes of flavonoids is

* Fax: +57-1-3165220.

E-mail address: jumarva@ciencias.unal.edu.co (J.C. Martínez Valderrama).

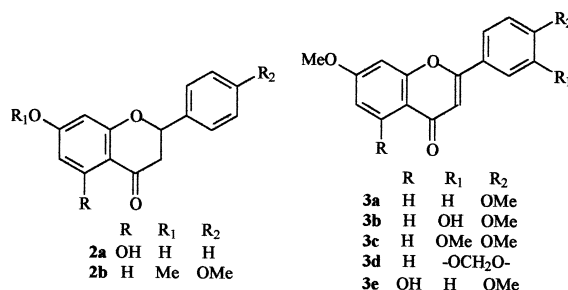
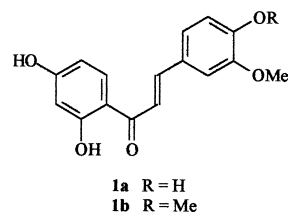
typical of some genera, and can be used to establish chemotaxonomic relationships.

2. Results

The flavonoids are compounds made up of two phenolic nuclei connected by three carbon units regarded as C₆–C₃–C₆. The different types of flavonoids vary in the oxidation state of the connecting C₃ moiety, which also determines the properties of the flavanoid. Types of flavonoids that have been found in the Myristicaceae species include chalcones, flavanones, flavones, dihydroflavonols, flavonols, flavans, flavan-3-ols, 1,3-diarylpropanes (virolanones), 1,3-diaryl-2-propanols (virolanols), dihydrochalcones, isoflavones and pterocarpanes. A biogenetic correlation among the Myristicaceae for all these types of flavonoids is shown in Chart 1.

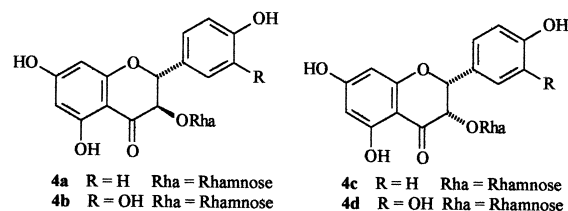
2.1. Chalcones (1)

Two chalcones, differentiated only by the hydroxyl or methyl groups at C-4, have been isolated and identified solely from *Iryanthera polyneura* (Almeida et al., 1979), and correspond to **1a** and **1b**.



2.2. Flavanones (2), flavones (3), dihydroflavonols (4) and flavonols (5)

Only two flavanones have been isolated from Myristicaceae species. One of them, **2a** (pinocembrin), with a phloroglucinol-type A ring, was isolated from *Iryanthera polyneura* wood (Almeida et al. 1979), and from *Virola surinamensis* twigs (Lopes et al., 1998) and roots (Lopes et al., 1999). The other, **2b**, with a resorcinol-type A ring, was isolated from *Virola carinata* wood (Gottlieb et al.,



1976). The absolute configuration at C-2 of these compounds is not known. Of the 315 flavones reported by Wollenweber (1993) which exhibited simple substitutions by hydroxyl, methoxyl and/or methylenedioxy groups, only five have been isolated, from four Myristicaceae species: **3a** from fruit (Cavalcante et al., 1985) and leaves (Daza, 1999) of *Virola flexuosa*; **3a**, **3b**, **3c** and **3d** from the leaves, flowers and fruit (seeds) of *Virola venosa* (Kato, et al., 1992); **3b** (tithonine) from *Virola surinamensis* seeds

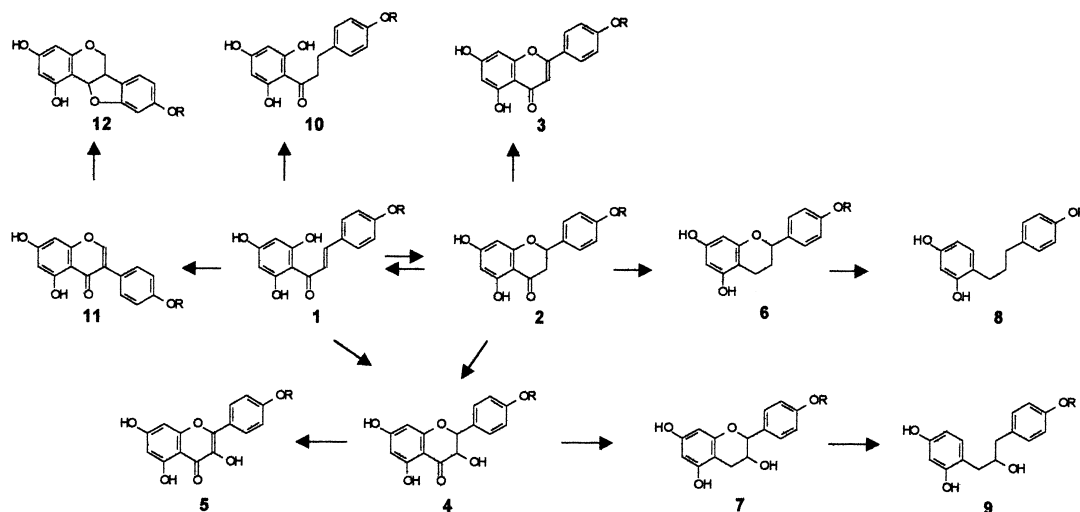
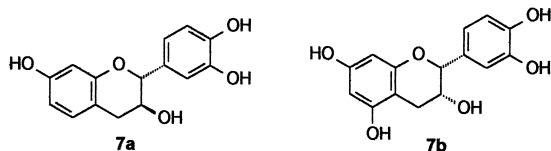
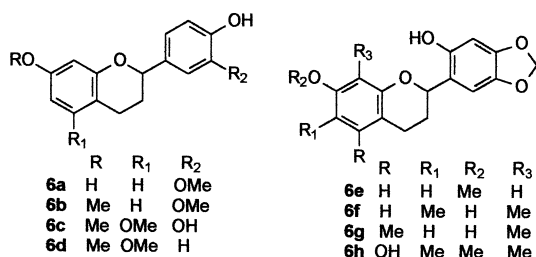
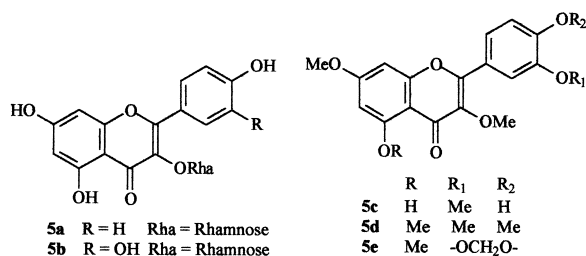


Chart 1. Biogenetic correlation of flavonoids isolated from Myristicaceae. Adapted from Gottlieb (1979).

(Blumental et al., 1997); and **3a**, **3b**, **3c** and **3e** from the leaves of *Virola michelii* (Santos et al., 1996).

Four 3-*O*- α -L-rhamnosyldihydroflavonols were identified from the inflorescences of *Iryanthera sagotiana* (Silva et al., 1997): **4a** (engeletin), **4b** (astilbin), **4c** (isoengeletin) and **4d** (isoastilbin), and two 3-*O*-rhamnosylflavonols **5a** (afzelin) and **5b** (quercitrin). **4a**, **4b**, **5a**, **5b** were isolated from the leaves of *I. sagotiana* (Silva et al., 1997). From the leaves of the *Otoba parvifolia*, three flavonols with C3-*O*-methyl groups (**5c**, **5d** and **5e**) were reported (Devia and Martínez, 1999).



2.3. Flavans (6) and flavan-3-ol (7)

The flavans and flavan-3-ols, though not very abundant in nature, serve as monomer units of proanthocyanidins; in the case of the Myristicaceae, they seem to be the precursors of virolanes and virolanol.

Eight flavans have been identified in Myristicaceae species (**6a–6h**), and all have been isolated from wood: **6a** from *Iryanthera elliptica* (Braz Filho et al., 1980b) and *Knema austrosiamensis* (Gonzales et al., 1993); **6b** from *Virola calophylloidea* (Martínez and Cuca, 1987); **6c** from *Iryanthera coriacea* (Franca et al., 1974; de Lima et al., 1975); **6a**, **6d** from *Iryanthera grandis* (Díaz and de Díaz, 1986); **6e** from *Iryanthera juruensis* (Franca et al., 1974); and **6f**, **6g** and **6h** from *Iryanthera laevis* (Braz Filho et al., 1980a). Compounds **6a–6d** are indicated to

exist as racemates; the stereochemistry of **6f**, **6g** and **6h** is not known.

Only two flavan-3-ols have been reported, corresponding to **7a** ((–)fisetinidol) isolated from *Virola elongata* wood (Kijjoa et al., 1981), and **7b** ((–)epicatechin) from *Myristica fragrans* seeds (Gopalarkishnan and Mathew, 1983).

2.4. 1,3-Diarylpropanes (8) and 1,3-diaryl-2-propanols (9)

The presence of these classes of compounds is limited, with very few exceptions, to species of the Myristicaceae; since the first analogues were found in species of the *Virola* genus, they were named virolanes and virolanol (Braz Filho et al., 1973). Fifteen virolanes (**8a–8o**) and three virolanols (**9a–9c**) in the genera *Virola*, *Iryanthera* and *Knema* (Table 1) have been reported. All have been isolated from wood, as is common with the flavans and flavan-3-ols that are considered to be their biogenetics precursors. The stereochemistry of virolanols has been variously assigned as 2R (Kijjoa et al., 1981) and 2S (Fernandez et al., 1980).

The 1,3-diarylpropanes are used to prevent the growth of malaria mosquito larvae (Jurd, 1977) and the 1,3-diaryl-2-propanols inhibit electrically-induced contractions in smooth muscle (Hagos et al., 1987). Two structurally modified compounds of the 1,3-diarylpro-

Table 1
Distribution of 1,3-diarylpropanes and 1,3-diaryl-2-propanols in the Myristicaceae

Source	Compounds	References
<i>V. caducifolia</i>	8g , 9a	Braz Filho et al., 1976b
<i>V. calophylloidea</i>	8c	Martínez and Cuca, 1987
<i>V. divergens</i>	8g , 9a	Gottlieb et al., 1973; Fernandez et al., 1980
<i>V. elongata</i>	8g , 9a , 9b , 9c	Kijjoa et al., 1981
<i>V. flexuosa</i>	8g , 9a	Braz Filho et al., 1977
<i>V. guggenheimii</i>	8g , 9a	Fernandez et al., 1980
<i>V. melinonii</i>	8g , 9a	Gottlieb et al., 1973
<i>V. minutiflora</i>	8g	Kijjoa et al., 1981
<i>V. multinervia</i>	8a	Braz Filho et al., 1976a
	8g , 9a	Braz Filho et al., 1973
<i>V. pavonis</i>	8g , 9a	Gottlieb et al., 1973
<i>V. sebifera</i>	8a , 9b , 9c	Von Rotz et al., 1990
<i>V. surinamensis</i>	8g , 9a	Gottlieb et al., 1973
<i>V. venosa</i>	8g , 9a	Gottlieb et al., 1973
<i>I. coriacea</i>	8b , 8f , 8g , 8h , 8i , 8m	de Lima et al., 1975
<i>I. elliptica</i>	8b	Braz Filho et al., 1980a
<i>I. grandis</i>	8e , 8f , 8h , 8j , 8o	Díaz and de Díaz, 1986
<i>I. laevis</i>	8h , 8j , 8k , 8m	Braz Filho et al., 1980
<i>I. paraensi</i>	8b , 8f , 8g , 8h , 8j	Da Silva et al., 1984
<i>I. polyneura</i>	8f , 8j	Almeida et al., 1979
<i>I. tricornis</i>	8b , 8f , 8h	Da Silva et al., 1984; Salazar et al., 1988
<i>I. ulei</i>	8h , 8j , 8l , 8m , 8n	Conserva et al., 1990a
<i>Knema austrosiamensis</i>	8d	Gonzales et al., 1993

pane-type have been isolated: virolaflorin (**8p**) isolated from *Virola minutiflora* wood (Kijjoa et al., 1981) and spiroelliptin (**8q**) isolated from *Iryanthera elliptica* wood (Braz Filho et al., 1980b).

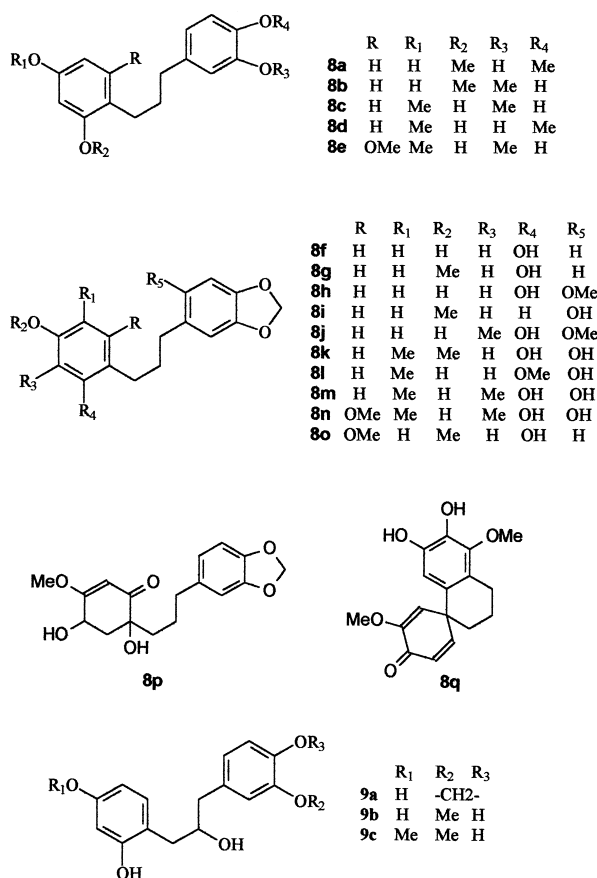
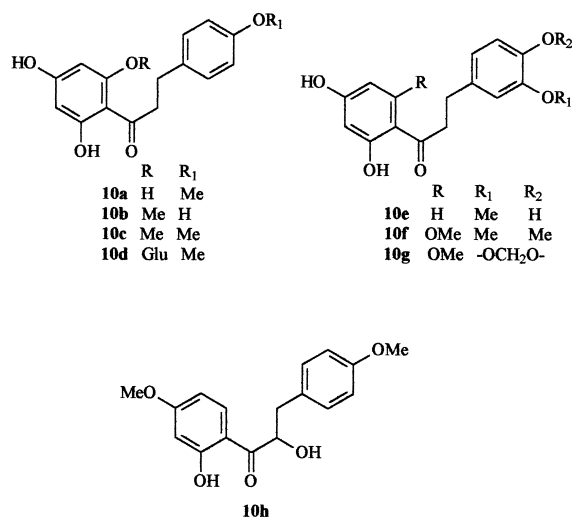


Table 2

Distribution of dihydrochalcones in the Myristicaceae

Source	Compounds	References
<i>I. grandis</i> , fruits	10c	Vieira et al., 1983; Silva et al., 1995
<i>I. laevis</i> , bark	10c, 10f, 10g	Martínez and Cuca, 1989
fruits	10a, 10c	Garzón et al., 1987
wood	10c	Braz Filho et al., 1980
<i>I. paraensis</i> , bark	10c	Conserva et al., 1990b
<i>I. polyneura</i> , wood	10e	Almeida et al., 1979
<i>I. sagotiana</i> , bark	10c, 10g	Kawanishi et al., 1990
inflorescence	10a, 10b, 10c, 10d	Silva et al., 1997
leaves	10b, 10c	Silva et al., 1997
<i>I. ulei</i> , bark	10f	Conserva et al., 1990a
wood	10c	Conserva et al., 1990a
<i>V. Calophylloidea</i> , wood	10h	Martínez and Cuca, 1987
<i>V. surinmensis</i> , twigs	10h	Lopes et al., 1998
roots	10h	Lopes et al., 1999
<i>V. carinata</i> , bark	10h	Benavides, 1999

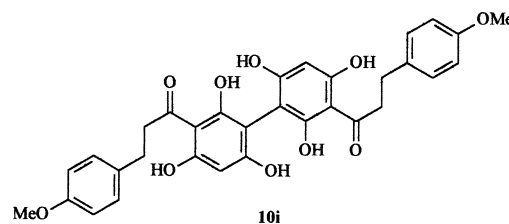


2.5. Dihydrochalcones (10)

Seven simple dihydrochalcones (**10a–10g**) from different parts of the plant (bark, inflorescences, fruit, leaves and wood) have been reported; they are very common in species of the *Iryanthera* genus (Table 2). The genus *Virola* has thus far yielded only the (±)-α-hydroxydihydrochalcone **10h**.

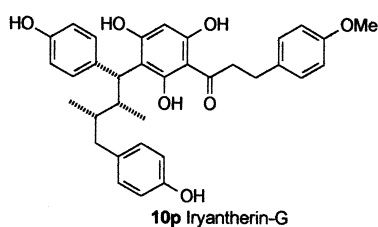
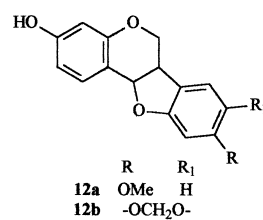
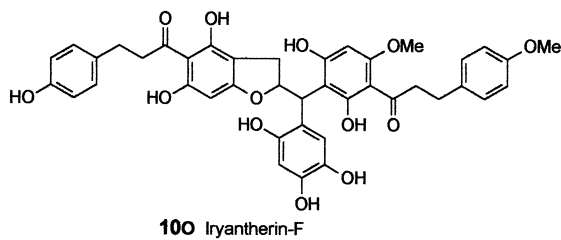
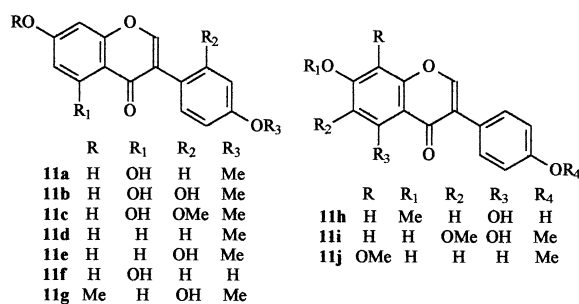
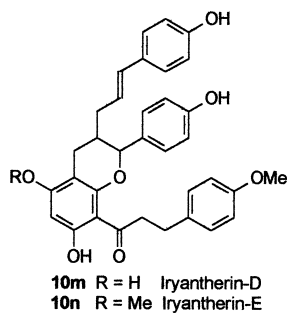
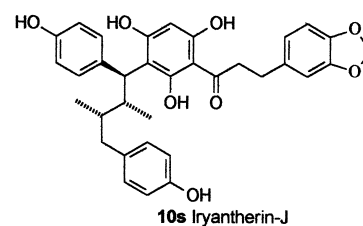
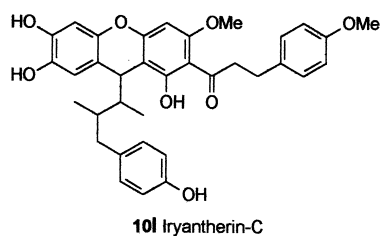
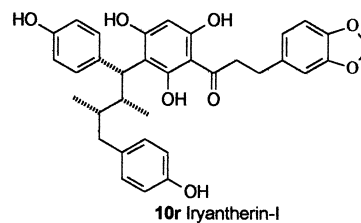
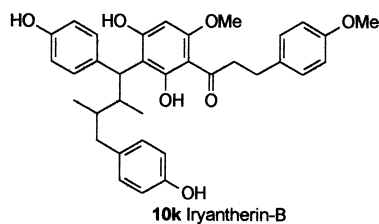
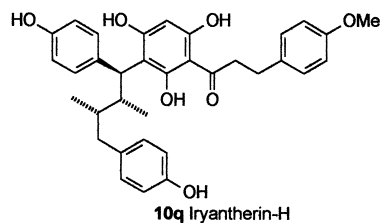
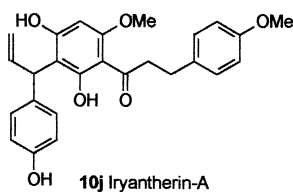
The first biflavonoid (**10i**) reported from the Myristicaceae family was isolated from the leaves of *Iryanthera sagotiana* (Silva et al., 1997), and is the second dimer of dihydrochalcone isolated as a natural product.

A complex group of dihydrochalcones, designated Iryantherins, has thus far been isolated exclusively from *Iryanthera* species: **10j**, **10k**, **10l**, **10m** and **10n** from *I. laevis* (Garzón et al., 1987; Martínez and Cuca, 1989; Conserva et al., 1990b); **10k**, **10m**, **10n** and **10o** from *I. ulei* (Conserva et al., 1990b); **10o** from *I. paraensis* (Conserva et al., 1990b); and **10p**, **10q**, **10r** and **10s** from *I. grandis* (Silva et al., 1995). Only relative configurations are known for Iryantherins **10p** to **10s**, as indicated in the structures.



2.6. Isoflavones (11)

From the results of some of the first studies on Myristicaceae species (Braz Filho et al., 1976a, 1977) it was concluded that isoflavonoids were a characteristic of this family (Gottlieb, 1979), but further reports of this type of compound have only recently begun to appear (Devia and Martinez, 1999; Benavides, 1999). Ten isoflavonoids (**11a–11j**) have been isolated from seven species of Myristicaceae (Table 3).



2.7. Pterocarpan (12)

Two pterocarpan, namely (±)-3-demethylhomopterocarpin (medicarpin, **12a**) and (±)-maackiain (**12b**) (Braz Filho et al., 1977), have been reported, both from the leaves of *Osteophloeum platyspermum*.

Table 3
Distribution of isoflavonoids in the Myristicaceae

Source	Compounds	References
<i>V. caducifolia</i> , wood	11a, 11b, 11c	Braz Filho et al., 1976b
	11a, 11b, 11c, 11d, 11e	Braz Filho et al., 1977
<i>V. carinata</i> , bark	11a, 11d, 11e, 11f, 11h	Benavides, 1999
<i>V. multinervia</i> , wood	11b, 11c, 11d, 11e	Braz Filho et al., 1977
<i>V. surinamensis</i> , twigs	11a, 11b, 11e	Lopes et al., 1998
roots	11a, 11d, 11g	Lopes et al., 1999
seed	11i	Blumental et. al. 1997
<i>Knema glomerata</i> , bark	11j	Zeng et al., 1994
<i>Otoba parvifolia</i> , bark	11a, 11h	Devia and Martínez, 1999
<i>Pycnanthus angolensis</i> , wood	11e, 11g	Omobuwajo et al., 1992

3. Conclusion

The established occurrence of flavonoids in seven genera of Myristicaceae suggests that this class of metabolites is a general feature of this family. There are twelve types of flavonoids reported in Myristicaceae, among which the aromatic B ring (derivative from shikimic acid) is either oxygenated at C-4 or dioxygenated at C-3, C-4 but not trioxxygenated at C-3, C-4, C-5 as expected according to the phenylpropanoids such as myristicin (Forrest et al., 1972) and elemicin (Barata et al., 1978) isolated from this family. Finally, it is of note that the presence of 1,3-diarylpropanes and 1,3-diaryl-2-propanols is restricted to the wood of *Virola*, *Iryanthera* and *Knema* genus; further, the occurrence of dihydrochalcones is a chemotaxonomic characteristic of the *Iryanthera* genus, and the Iryantherins are unique to this family.

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