

SHORT COMMUNICATION

A LIGNAN AND ARYL ALKANONES FROM ROOT BARK OF *MYRISTICA CEYLANICA*

H.M.T. BANDARA HERATH

Institute of Fundamental Studies, Hantana Road, Kandy.

(Received: 26 June 1996; accepted: 03 January 1997)

Abstract : A lignan, rel-(8S,8'R)-dimethyl-(7S,7'R)-bis(3,4-methylenedioxyphenyl) tetrahydrofuran and four aryl alkanones, 1-(2',6'-dihydroxyphenyl)tetradecan-1-one, 1-(6'-hydroxy-2'-methoxyphenyl)-9-(4"-hydroxyphenyl)nonan-1-one, 1-(2',6'-dihydroxyphenyl)-9-(4"-hydroxyphenyl)nonan-1-one and 1-(2',6'-dihydroxyphenyl)-9-(3",4"-dihydroxyphenyl)nonan-1-one were isolated from the hot dichloromethane extract of the root bark of *Myristica ceylanica*. Although this is the first report of these compounds from *M. ceylanica*, they have been isolated from other *Myristica* species. Their structures were established by comparison with previously reported physical data and by chemical conversions.

Key words: Aryl alkanone, lignan, *Myristica ceylanica*, Myristicaceae.

INTRODUCTION

The family Myristicaceae which consists of nineteen genera and about four hundred and forty species is exclusively tropical and found mainly in the lowland rain forests in the Asian tropics, tropical America, Africa and Madagascar.¹ However the genus *Myristica* which consists of about eighty species is found only in the Asian tropics.¹ *M. ceylanica* is the only endemic among the three *Myristica* species found in Sri Lanka. Of the other two species, *M. dactyloides* is also found in South India and *M. fragrans* in almost all the South-East Asian countries.^{1,2} Although the chemical constituents of *M. fragrans* have been studied extensively since the beginning of the 20th century due to their pharmacological properties, the other two Sri Lankan species have been relatively less explored. Previous studies on *M. dactyloides* include reports on the presence of myoinositol in the hot methanol extract³ and eight aryl alkanones and two lignans in the hot dichloromethane extract.^{4,5} There has been no previous report on the chemical investigation of *M. ceylanica* (S - Malaboda) which is a large tree found in the lowland rain forests of Sri Lanka. Because of the great interest in the chemistry of Sri Lankan Myristicaceae, we investigated the root bark extract of *M. ceylanica*. Herein we describe the isolation of a lignan, rel-(8S,8'R)-dimethyl-(7S,7'R)-bis(3,4-methylenedioxyphenyl) tetrahydrofuran (**1**), and four aryl alkanones 1-(2',6'-dihydroxyphenyl) tetradecan-1-one(**2**), 1-(6'-hydroxy-2'- methoxyphenyl) - 9 - (4" - hydroxyphenyl) nonan - 1 - one (**3**), 1 - (2',6' - dihydroxyphenyl) - 9 -

(4''-hydroxyphenyl)nonan-1-one(4) and 1-(2',6'-dihydroxyphenyl)-9-(3'',4''-dihydroxyphenyl) nonan-1-one(5) from the hot dichloromethane extract of the root bark.

METHODS AND MATERIALS

Melting points were recorded on a Kofler hot-stage apparatus. Identities of compounds were established by mmp, co-TLC, IR, NMR and MS comparisons. Optical rotations were measured using a Perkin Elmer 241 polarimeter at 22°C. ¹H and ¹³C-NMR spectra were recorded respectively on a Varian Gemini spectrometer at 200 MHz and 50 MHz and the ¹H-NMR spectra of methylated products on a Varian T-60 spectrometer. IR spectra were recorded on a Shimadzu IR-460 instrument in KBr discs unless otherwise stated and MS spectra on a Shimadzu QP-1000A spectrometer. Prep. TLC was carried out on Merck Kieselgel 60 F₂₅₄. Flash and medium pressure column chromatography were carried out on Merck Kieselgel 60 (230-400 mesh ASTM).

Plant material: *Myristica ceylanica* was collected from the Kanneliya forest reserve in the southern province of Sri Lanka and identified by comparison with the Herbarium specimen (2367, collected by Jayasooriya, & Kostermans on 26th July 1976) at the National Herbarium, Royal Botanic Gardens, Peradeniya, Sri Lanka.

Extraction and isolation: The dried, powdered root bark of *M. ceylanica* (450 g) was extracted successively with hot hexane, hot CH₂Cl₂ and hot MeOH. The hot CH₂Cl₂ extract (10 g) was flash chromatographed over silica gel and gradient eluted with hexane - CH₂Cl₂.

Compound 1: Elution with CH₂Cl₂ - hexane (1:6) by prep. TLC to give rel-(8S,8'R)-dimethyl-(7S,7'R)-bis(3,4-methylenedioxyphenyl) tetrahydrofuran (1, 62 mg) as a colourless oil; [α]_D: 0° (lit. [5]. [α]_D: 0°).

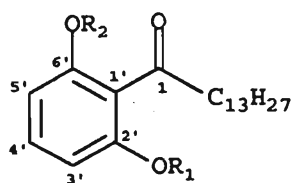
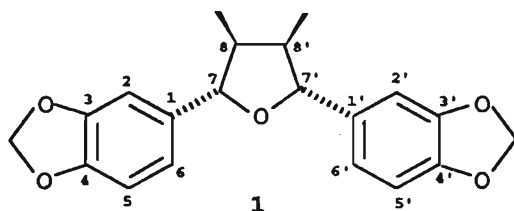
Compound 2 and 3: The phenolic extract from the fraction eluting with CH₂Cl₂ - hexane (1:4) gave, on further purification by prep. TLC 1-(2',6'-dihydroxyphenyl) tetradecan-1-one (2, 143 mg), m.p. 92-94°C (lit.[4] m.p.91-92°C), while that from the fraction eluting with CH₂Cl₂ - hexane (1:4 to 1:3) on purification by flash chromatography and recrystallization gave 1-(6'-hydroxy-2'-methoxyphenyl)-9-(4''-hydroxyphenyl)nonan-1-one, (3, 560 mg), m.p. 68°C (lit. [4] m.p. 65-66°C).

Compound 4 and 5: Elution with CH₂Cl₂ - hexane (2:3) gave 1-(2',6'-dihydroxy phenyl)-9-(4''-hydroxyphenyl) nonan-1-one (4, 342 mg) m.p. 105-106°C (lit. [4]m.p. 105-106°C), while elution with MeOH-CH₂Cl₂ (1:19) gave on prep. TLC, 1-(2',6'-dihydroxyphenyl)-9-(3'',4''-dihydroxyphenyl) nonan-1-one (5, 43 mg), m.p. 124-126°C (lit. [6] m.p. 123-124°C).

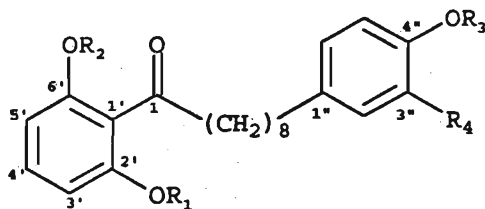
Methylation of compounds 2, 3 and 4: Methylation of **2** (50 mg) with K_2CO_3 (60 mg) and MeI (4 ml) in anhydrous acetone (10 ml) at 26°C for 2 h gave on separation with prep. TLC, the less polar 1-(6'-hydroxy-2'-methoxyphenyl)tetradecan-1-one (**2a**, 23 mg) mp 52-54°C (lit. [4] m.p. 51-52°C) and the more polar 1-(2',6'-dimethoxyphenyl)tetradecan-1-one (**2b**, 12 mg), whose spectral data were identical with those reported previously.⁴

Methylation of **3** (30 mg), with K_2CO_3 (40, mg) in anhydrous acetone (10 ml) at 26°C for 6 h gave, on purification with prep. TLC, 1-(2',6'-dimethoxyphenyl)-9-(4'-methoxyphenyl)nonan-1-one (**3a**, 17 mg), whose spectral data were identical with those reported previously.⁴

Methylation of **4** (50 mg) under the same conditions gave compounds **3** and **3a**, identical with those obtained above.



	R ₁	R ₂
2	H	H
2a	CH ₃	H
2b	CH ₃	CH ₃



	R ₁	R ₂	R ₃	R ₄
3	CH ₃	H	H	H
4	H	H	H	H
5	H	H	H	OH
3a	CH ₃	CH ₃	CH ₃	H

RESULTS AND DISCUSSION

The structural elucidation of compound **1** has been previously described.⁵ The configurations of the two methyl and the two aryl groups in the compound were confirmed by comparing the spectra of **1** with that of rel-(8S,8R')-dimethyl-(7S,7'R)-bis(4-hydroxy-3-methoxyphenyl)tetrahydrofuran from *Jatropha grossidentata*.⁷ Compounds **2**, **3**, and **4** have been previously isolated from *M. dactyloides*⁴ and their structures were confirmed by comparison with authentic samples and also by the methylation reactions described above. The structure of compound **5** was established by comparison of its spectral data with those of 1-(2',6'-dihydroxyphenyl)-9-(3'',4''-dihydroxyphenyl)nonan-1-one, previously isolated from *M. malabarica*.

Acknowledgements

I thank Dr. D. Bergenthal at the Institute of Pharmaceutical Chemistry, University of Muenster, Germany for providing 200 MHz ¹H-NMR and 50 MHz ¹³C-NMR spectral data and N. Chandrasiri of the Department of Chemistry, University of Peradeniya for 60 MHz spectral data of the methylated products, S. Ekanayake for collection and identification of plant material, and C. Dissanayake for the technical support.

References

1. Philcox D. (1996). In: *A Revised handbook to the flora of Ceylon*. (Eds. M.D. Dassanayake & W.D. Clayton) Vol. XI (in press), Amerind Publishing Co. (Pvt). Ltd., New Delhi, India.
2. Jayaweera D.M.A. (1982). *Medicinal plants used in Ceylon* (Part IV) pp. 107, The National Science Council, Colombo, Sri Lanka.
3. Tillekaratne L.M.V., Jayamanne D.T. & Weerasooriya K.D.V. (1981). Chemical constituents of *Myristica dactyloides*. *Journal of the National Science Council of Sri Lanka* **9**: 251-253.
4. Kumar N.S., Herath H.M.T.B. & Karunaratne V. (1988). Arylalkanones from *Myristica dactyloides*. *Phytochemistry* **27**: 465-467.
5. Herath H.M.T.B & Priyadarshani A.M.A. (1996). Two lignans and an arylalkanone from *Myristica dactyloides*. *Phytochemistry* **42**: 1439-1442.
6. Purushothaman K.K., Sarada A. & Connolly J.D. (1977). Malabaricones A-D, novel diarylnonanoids from *Myristica malabarica* Lam (Myristicaceae). *Journal of Chemical Society. Perkin Trans* **1(5)**: 587-588.
7. Schmeda-Hirschmann G., Tschirz F. & Jakupovic J. (1992). Diterpenes and a lignan from *Jatropha grossidentata*. *Phytochemistry* **31**: 1731-1735.