JANGOMOLIDE, A NOVEL LIMONOID FROM FLACOURTIA JANGOMAS

Jamal Ahmad, Kausar Wizarat, K. M. Shamsuddin, Asif Zaman and Joseph D. Connolly*

Department of Research in Unani Medicine, Aligarh Muslim University, Aligarh 202001, India; *Department of Chemistry, University of Glasgow, Glasgow G12 8QQ, Scotland

(Received 13 September 1983)

Key Word Index—Flacourtia jangomas; Flacourtiaceae; structure; limonoids; jangomolide.

Abstract—Jangomolide, a novel limonoid has been isolated, together with limonin, from Flacourtia jangomas.

INTRODUCTION

Flacourtia jangomas (Lour.) Raeusch (syn. F. cataphracta Roxb.) is a small tree which grows in northern India near the Nepal border. Its dried leaves are used locally as a treatment for asthma. Extraction of the dried stem and bark with light petroleum yielded ostruthin (1). The acetone and ethanol extracts afforded a crystalline mixture of two limonoids separable by preparative TLC. The more polar compound was readily identified as limonin (2). The less polar component was a novel limonoid, jangomolide, which was assigned structure 3 on the following evidence.

RESULTS AND DISCUSSION

Jangomolide, $C_{26}H_{28}O_8$, mp 282–288° (ex chloroform-ether), v_{max} (CCl₄) 1773 cm⁻¹, like limonin, has four tertiary methyl groups [δ_H 1.36, 1.28 (6H), 1.16; δ_C 31.5, 24.7, 20.0, 15.5], a cyclohexanone (δ_C 209.1), a β-substituted furan [δ_H 7.41 (m, H-21 and H-23) and 6.33 (m, H-22); δ_C 143.2 (C-23), 141.1 (C-21), 119.8 (C-20) and 109.6

(C-22)] and a ring D epoxy-lactone [δ_H 3.99 (s, H-15), 5.53 (s, H-17); δ_C 67.2 (C-14), 54.1 (C-15), 166.8 (C-16) and 77.8 (C-17)]. In addition the ¹H and ¹⁷C spectra indicate the presence of an α,β -unsaturated lactone [δ_H 6.54 and 6.14 (both d, J = 9.6 Hz, H-1 and H-2); δ_C 150.8 (d, C-1), 118.8 (d, C-2), 160.41 (s, C-3)], a tertiary carbon bearing oxygen [δ_C 86.3 (s, C-4)] and a secondary acetal carbon [δ_H 6.06 (s, H-19); δ_C 104.0 (d, C-19)]. These features are obviously associated with ring A of the tetranortriterpenoid skeleton and can readily be assembled to give structure 3 for jangomolide. The R configuration of C-19 follows from the observation of a small ⁴J coupling between H-19 and H-1 since only this configuration permits the correct geometrical relationship for such a coupling.

This is the first report of the presence of limonoids in a member of the Flacourtiaceae. Normally limonoids are found in the Meliaceae, the Rutaceae and the Cneoraceae. The plant material used in this investigation was identified at the Royal Botanical Gardens, Kew, as F. jangomas and jangomolide was isolated from the reference sample. The Flacourtiaceae are well-known for their seed oils, containing cyclopentanoid fatty acids. Further work identified

1270 J. Ahmad et al.

xanthones and friedelane triterpenoids [1]. The isolation of limonoids from a member of this family raises interesting chemotaxonomic questions, the significance of which must await further study.

EXPERIMENTAL

Plant material This was collected near Gorakhpur in Northern India and a reference specimen has been deposited in the Dept. of Chemistry, University of Glasgow.

Isolation. Air dried stem and bark of F. jangomas (5 kg) was extracted with petrol at room temp, for a week. On standing at ice temp., the cone soln afforded a yellow solid ppt (2 g) which was purified by chromatography over silica gel and crystallisation from C_6H_6 -petrol to give ostruthin (1) mp 116°, identified by its spectroscopic properties

The defatted plant material was extracted successively with EtOH and Me_2CO and the resulting extract, after evaporation of solvent, partitioned between EtOAc and H_2O . Removal of the EtOAc left a brown solid (700 mg) which was chromatographed over silica gel Elution with C_6H_6 -EtOAc and crystallisation

from CHCl₃-petrol gave a crystalline material (300 mg), mp 284° which was shown to be a mixture of two components by analytical TLC. Preparative TLC of a sample (100 mg) of this mixture yielded limonin (2, 40 mg), the more polar compound, identical with an authentic specimen The less polar compound was crystallized from CHCl₃-Et₂O to give jangomolide (3) (40 mg) mp 282–288° [$\delta_{\rm C}$ 38.4 (s, C-13), 49.2 and 49.3 (both s, C-10 and C-8), 41.6 (d, C-9), 53 2 (d, C-5), 19.0 (t, C-11), 29.0 (t, C-12) and 37.5 (t, C-6)]. [Found. m/z 453 1540 [M – 15]⁺. C₂₅H₂₅O₈ requires m/z 453.1549.]

Acknowledgements—The Indian authors wish to thank C.C.R.U M., Ministry of Health, Government of India, for financial assistance Thanks are also due to Professor D. L Dreyer for valuable discussions and Dr B Mathew, Royal Botanic Gardens for identification of the plant material.

REFERENCE

 Gunasekera, S. P., Sultanbawa, M. U. S. and Balasubramanian, S. (1977) Phytochemistry 16, 788.