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BIFLAVONES FROM THE LEAVES OF *ARAUCARIA ARAUCANA*

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A number of biflavones have been reported from the leaves of *Araucaria bidwilli* (1-3), *Araucaria cookii* (3-7), *Araucaria cunninghamii* (3,6,8,9), *Araucaria excelsa* (10,11), and *Araucaria rulei* (1). The chemotaxonomic significance of the biflavones in the genus *Araucaria* (3) and their anticancer activity (N.U. Khan, N. Parveen, M. Parveen, and H.M. Taufeeq, unpublished results) prompted us to investigate *Araucaria araucana* (Molina) K. Kock (Araucariaceae). In the present communication we report the occurrence of several biflavones isolated by the method of Khan *et al.* (3). The major leaf constituents identified were 7-*O*-methylgathisflavone, 7ⁿ-*O*-methylamentoflavone, and 7,7ⁿ-di-*O*-methylcupressuflavone.

The minor constituents, di-*O*-methylgathisflavone, di-*O*-methylamentoflavone, tri-*O*-methylgathisflavone, tri-*O*-methylamentoflavone, and tri-*O*-methylcupressuflavone were tentatively identified based on their R_f values and characteristic colors in uv light with authentic samples (3,4).

EXPERIMENTAL

PLANT MATERIAL.—*A. araucana* was collected from Lylod Botanical Garden, Darjeeling, in March 1984 and identified by Dr. W. Husain, Reader, Department of Botany, A.M.U., Aligarh. A voucher specimen was submitted to the A.M.U. Herbarium, Aligarh (Voucher No. Husain-49601).

ISOLATION AND IDENTIFICATION.—Dried and powdered leaf material of *A. araucana* (1 kg), after being defatted with light petrol, was extracted with Me₂CO. The Me₂CO extract was concentrated, and the residue was refluxed with light petrol, C₆H₆, and CHCl₃, treated with boiling H₂O, and filtered. The yellow, solid, undissolved residue (2.5 g) yielded six chromatographically homogeneous fractions, AA-I to AA-VI, after development on a Si gel column and subsequent preparative tlc (Si gel, C₆H₆-pyridine-

HCOOH, 36:9:5). AA-IV was further separated to AA-IVa and AA-IVb by preparative tlc (Si gel, C₆H₆-pyridine-HCOOH, 40:10:2). AA-I, AA-II, and AA-IVb were identified as 7-O-methylagathisflavone, 7"-O-methylamentoflavone, and 7,7"-di-O-methylcupressuflavone, respectively, by comparison of their mp, R_f values, and ¹H nmr with authentic samples. AA-III, AA-IVa, AA-V, and AA-VI were tentatively identified by comparison of these compounds and their permethylated derivatives with authentic samples (1,3,4,9), based on their R_f value and characteristic colors in uv light. AA-III and AA-V were comparable with 7,7"-di-O-methylagathisflavone (3) and 4',7,7"-tri-O-methylagathisflavone (4), respectively, and their permethylated derivatives were identical with the agathisflavone hexamethyl ether (3). Therefore, AA-III and AA-V were identified as di- and tri-O-methylagathisflavone, respectively. AA-IVa, identified as di-O-methylamentoflavone, compared with 4',7"-di-O-methylamentoflavone (3) and its permethyl ether was identical with amentoflavone hexamethyl ether (3). AA-V, an inseparable isomeric mixture of the trimethyl ethers of amentoflavone and cupressuflavone, was comparable with kayaflavone and 4',7,7"-tri-O-methylcupressuflavone (1,3), and on permethylation it gave a mixture of hexamethyl ethers of amentoflavone and cupressuflavone (3). Full details are available upon request to the senior author.

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