Isolation of 3β, 16α, 17-trihydroxyphyllocladane from leaves of Callicarpa macrophylla Vahl.

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Abstract

A new kaurane diterpene isolated from chloroform extract of leaves of *Callicarpa macrophylla* and it has been characterized as 3*β*,16*α*,17-trihydroxyphyllocladane along with calliterpenone and its monoacetate.

Keywords: *Callicarpa macrophylla*; *Verbenaceae; calliterpenane;* 3*β*,16*α*,17-trihydroxyphyllocladane.

Introduction

Callicarpa macrophylla Vahl. (Family: *Verbenaceae*) is an important but non famous medicinal plant at Himalaya and it is commonly known as Priyangoo or Daya. It is a perennial, deciduous shrub attaining 2.5 m in height. Many kinds of diseases can be cured by this plant. The bark can be used to heal cut and wounds. Seeds and roots can be used for curing digestion and leaves can be used in rheumatism. In addition, fruits can be used for healing blisters and boils. Extracts of this plant also show anti-inflammatory, antifungal and antibacterial activities [1-8].

In addition to previous work [9], a new compound was isolated from this plant. The compound was characterized as 3*β*,16*α*,17-trihydroxyphyllocladane.

Material and methods

Fresh leaves of *Callicarpa macrophylla* Vahl. were procured from the Central Institute of Medicinal and Aromatic Plants (CIMAP) Lucknow (U.P.) in October 2009. The plant was identified by a Taxonomist of the center and a specimen was kept for record.

The air dried plant material was coarsely powered (1.5 kg) and then sequential extracted with petroleum ether (60 – 80 ^{0}C), chloroform and methanol by the soxhlet apparatus (5 times x 1 L. each). The fractions of each extracts were mixed together and excess solvent was evaporated under reduced pressure. Among these extracts, only chloroform extract was considered for further examination. The semisolid brownish mass (3 g) obtained from chloroform extract was dissolved in small amount of

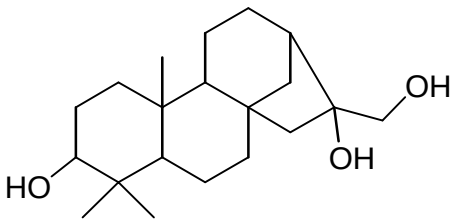
chloroform and was mixed with 3 g of silica gel. The slurry was loaded on a column of silica gel (60 g) and eluted with petroleum ether, benzene, chloroform, ethyl acetate, methanol and their mixtures of different proportions of increasing polarities. Several fractions were obtained which were monitored by TLC and the fraction showing single spot on TLC were combined together.

The compound was obtained as crystalline solid by eluting with benzene – chloroform (1:9), m.p., 164 -166 ^{0}C and was characterized as 3β, 16α, 17-trihydroxyphyllocladane by comparing spectral data
^{1}H NMR($CDCl_3$) δ 3.64 (2H, H-17), δ 3.19(1H, H-3), δ 0.99 – δ 1.10 (9H, 3x-CH_3); ^{13}C NMR ($CDCl_3$) δ 29.2(C-1), δ 28.8(C-2), δ 79.6(C-3), δ 36.1(C-4), δ 49.2(C-5), δ 20.1(C-6), δ 32.3(C-7), δ 29.9(C-8), δ 51.0(C-9), δ 30.3(C-10), δ 20.5(C-11), δ 21.8(C-12), δ 40.5(C-13), δ 32.3(C-14), δ 40.8(C-15), δ 72.2(C-16), δ 76.8(C-17), δ 19.1(C-18), δ 20.5(C-19), δ 23.2(C-20).

Results and discussions

The compound was obtained by eluting the column with benzene – chloroform (1:9). It did not show any absorption band in IR spectrum corresponding to carbonyl function but had clearly indicated band at 3410 cm^{-1} for hydroxyl group. The molecular ion peak in mass spectrum at m/z=322 was consistent with the molecular formula $C_{20}H_{34}O_3$. The ^{1}H NMR spectrum had a two proton multiplet at δ 3.64 and another one proton gave broad doublet at δ 3.19 which can be assigned to hydroxy methylene (H – 17) and hydroxymethine (H – 3) protons respectively. Three tertiary methyl protons resonate at δ 0.99 – δ 1.10. The ^{13}C NMR spectrum indicated the presence of three methyl, nine methylene, four methine and four non – protonated carbon atom resonances but did not show any resonances regarding the carbonyl and olefinic functions further supported the phyllocladane structures. The downfield signals at δ 79.6, δ 76.8 and δ 72.2 were clearly assignable to hydroxy substituted carbon C-3, C-17 and C-16 respectively. The three tertiary methyl groups resonated at δ 19.1 – δ 28.8.

On the basis of the above observation, the compound was suggested to be tetracyclic diterpene of phyllocladane skeleton with three hydroxy groups attached at C-3, C-16 and C-17.
The compound was characterized as 3*β*,16*α*, 17-trihydroxyphyllocladane and this is the first time to be isolated from this plant.



3*β*,16*α*, 17-trihydroxyphyllocladane

References

1. R. N. Chopra; S. L. Nayar; I. C. Chopra. *Gloss Indian Med. Plant, CSIR,PID, N.Delhi*, **1956**.
2. P. S. Gupta; M. Sen; B. C. Pal. *J. Indian Chem. Soc.*, LIII, **1976**, 218 – 219.
3. A. K. Singh; A. K. Bhattachaary; G. Prasad; V. D. Sharma; K. C. Gupta. *Indian J. Microb.*, **1982**, 22(4), 286-288.
4. K. R. Kirtikar; B. D. Basu. Indian Medicinal Plants, *Dehradoon*, **1984**.
5. S. P. Ambasta; K. Ramachandran; K. Kashyapa; R. Chand. The useful plants of India, *CSIR, N. Delhi*, **1986**.
6. A. K. Sharma; K. S. Negi; D. C. Bhandari; H. Y. Shukla; S. K. Pareek. *ENVIS Bulletin,* **2004**, 12(1), 38-40.
7. W.P. Jones and D. Kinghorn , Curr. *Bioact. Compd.* **2008**, 4(1),15-32.
8. R. K. Verma; A. K. Singh; Pooja Srivastava; K. Shanker; A. Kalra; M. M. Gupta. *J. Liquid Chromatog*., **2009**, 32, 2437-2450.
9. V. K. Verma; N.U.Siddiqui; Mohd. Aslam. *Biosci. Biotech. Res. Asia*, **2010**, 7(2), 1047 – 1048.