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A New Kaurane Diterpene from the Leaves of *Callicarpa macrophylla* Vahl

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ABSTRACT

The chloroform extract of leaves of *Callicarpa macrophylla* Vahl., yielded a new compound 16α-hydroxy- 17-isopropylidino-3-oxo-phyllocladane along with calliterpenone and its monoacetate.

Key words: *Callicarpa macrophylla*, Verbenaceae, Chloroform extract 16α-hydroxy- 17-isopropylidino-3-oxo-phyllocladane.

INTRODUCTION

Plant *Callicarpa macrophylla* Vahl. (Family: Verbenaceae) is an important less known medicinal plant of the lower warm valleys of the Himalaya and is commonly known as Priyangoo or Daya. It is a perennial, deciduous shrub attaining 2.5mt. in hight. Essential oil obtained from different parts of Priyangoo through steam distillation, while no essential oil content was observed in the roots[1-5]. All The parts are important and are used to cure many diseases. The bark is used to heal cut and wounds. Seeds and roots are used for digestion and leaves are used in rheumatism. The fruits are used for blisters and boils. Various extracts of this plant have shown anti-inflammatory, antifungal, antimicrobial and antibacterial activities[6-10].

In continuation to our previous work[11], here we report a new compound isolated from the plant. The compound was characterized as 16α-hydroxy-17-isopropylidino-3-oxophyllocladane.

MATERIAL AND METHODS

The fresh leaves of *Callicarpa macrophylla* Vahl. (Family: Verbenaceae) were procured from the Central Institute of Medicinal and Aromatic Plants (CIMAP) Lucknow (U.P.) in Octuber 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 (60^0-80^0C), chloroform and methanol by the soxhlat apparatus (5 times x 1Lit. each).The fractions of each extract were mixed together and the excess of solvent was evaporated under reduced pressure. Out of these extracts only chloroform extract was considered for further examination. The semisolid brownish mass (3 gm) obtained from chloroform extract was dissolved in small amount of chloroform and was mixed with 3 gm of silica gel. The slurry was loaded on a column of silica gel (60 gm) and eluted with petroleum ether, benzene, chloroform, ethyl acetate, methanol and their mixtures of different proportions of increasing polarity. Several fractions were obtained which were monitored by TLC and the fraction showing single spot on TLC were combined together.

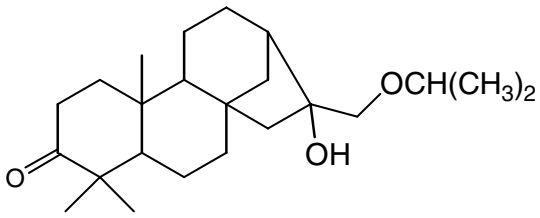
The compound was obtained as crystalline solid by eluting with benzene – chloroform (2:8), m.p.,162^0-164^0C and was characterized as16α-hydroxy- 17-isopropylidino-3-oxophyllocladane by comparing spectral data ^{1}H

NMR ($CDCl_3$) δ 4.01 & δ 3.98 (2H, d, oxymethylene protons H-17), δ 3.19(1H, oxymethine proton), δ 0.99 - δ 1.31 (15H, 5x-CH_3); ^{13}C NMR ($CDCl_3$) δ 32.0(C-1), δ3 3.6(C-2), δ 213.0(C-3), δ 50.1(C-4), δ 48.8(C-5), δ 21.9(C-6), δ 32.0(C-7), δ 31.8(C-8), δ 49.4(C-9), δ 29.4(C-10), δ 20.6(C-11), δ 22.8(C-12), δ 39.8(C-13), δ 32.5(C-14), δ 41.2(C-15), δ 74.9(C-16), δ 76.5(C-17), δ 69.0(C-18), δ 18.9(C-19), δ 20.1(C-20), δ 20.9(C-21), δ 23.6(C-22), δ 24.1(C-23)

RESULTS AND DISCUSSION

The compound was obtained as crystalline solid by eluting with benzene – chloroform (2:8). The I.R. spectrum of the compound showed the presence of hydroxyl group (3400 cm^{-1}) as well as carbonyl group (1700 cm^{-1}). The mass spectrum of the compound indicated the molecular ion peak at m/z=362 corresponding to molecular formula $C_{23}H_{38}O_3$, suggesting the existence of the five double bond equivalence. The disappearance of olefinic signals in ^1H and ^{13}C NMR spectra and appearance of resonances of number of rings and large number of methyl groups, the compound was suggested to be a saturated tetracyclic diterpene having kaurane skeleton of phyllocladane type. The ^1H NMR spectrum of the compound exhibited a pair of doublets at δ 3.98 and δ 4.01 reasonably assignable to an oxymethylene protons. The signal at δ 3.19 was due to oxymethine proton. The signal for five methyl groups resonates between δ 0.99 – δ 1.31. The structure was further justified by ^{13}C NMR spectral studies which showed twenty three carbon signals out of which five are for methyl, nine for methylene , four for methine and five for unprotonated carbon atoms. A down field signal appeared in ^{13}C NMR spectrum at δ 213.0 was assignable to carbonyl carbon C-3. The other downfield signals found at δ 76.5 was for an oxymethylene C-17, at δ 74.9 was due to oxysubstituted non protonic carbon C-16 and at ä 69.0 was due to isopropylene carbon. The signal for gem-dimethyl group appeared at δ 23.6. The rest of signal have resemblance with those of calliterpenone and resonates between δ 18.9-δ 50.1.

On the basis of these observations the structure of the compound was assigned as 16α-hydroxy- 17-isopropylidino-3-oxo-phyllocladane. This is the first report of the compound from this plant.



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