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Four diterpenes from *Callicarpa pedunculata*

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1. Subject and source

Callicarpa pedunculata R. Brown (Verbenaceae) is a small shrub widely distributed from southeastern and southern China to the Philippines and is very common in thickets at low altitudes (Cheung and Li, 1978). The aerial parts were collected in Guizhou Province, China, and a voucher specimen was deposited in the herbarium of the Kunming Institute of Botany, Chinese Academy of Sciences.(KUN No. 0300993).

2. Previous work

No previous phytochemical studies on *C. pedunculata* were reported. However, previous studies have revealed that *C. macrophylla* produced phyllocladane diterpenoids such as calliterpenone and its monoacetate, (Aziz Ahmad and Asif Zaman (1973); Chatterjee et al. (1972), and calliphylline, which is an isopimaradiene derivative (Sunil et al., 1994).

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3. Present study

Six kgs of dried and powdered leaves of *C. pedunculata* were extracted three times with EtOH under reflux. Removal of solvents under vacuum, gave a tar which was extracted with $CHCl_3$. Eighty grams of the $CHCl_3$ extracts was chromatographed over silica gel (200–300 mesh) and eluted with petrol, and petrol–EtOAc (10:1, 5:1, 2:1, 1:1), EtOAc, MeOH. The elutes were collected as 500ml fractions. The fractions eluted with petrol: EtOAc (5:1) were combined (12.5g) according to TLC and further purified by chromatography over Sephadex LH-20, reverse phase C-18 silica gel and recrystallization to yield compound **1** (40mg), compound **2** (25mg), compound **3** (22mg), compound **4** (35mg), respectively. These were identified by spectral analysis (IR, MS, ^{1}H NMR, ^{13}C NMR) and chemical evidence as 14α-hydroxy-7, 15-isopimaradien-18-oic acid (**1**) (Bruno et al., 1986); 16α,17-dihydroxy-3-oxophyllocladane (**2**) (Agrawal et al., 1995); 8,11,13,15-abietatraen-18-oic acid (**3**) (Tanaka et al., 1997); 6α-hydroxynidorellol (**4**) (Quijano et al., 1982) (all data were available on request).

4. Chemotaxonomic significance

Representatives of four classes of diterpenoids were isolated from *C. pedunculata*, whereas, only **2** had been previously isolated from this genus. Compound **1**, **3** and **4** have been reported from many other sources, such as **1** from *Salvia greggi* (Labiatae), **3** from *Larix kaempfer* (Pinaceae), **4** from *Stevia monardaedaefolia* (Compositae). This work is the first example of the co-occurrence of four types of diterpenoids in a single species of Verbenaceae. The diversity of diterpenoids in *C. pedunculata* is similar to that in some Labiatae species (Sandra et al., 2001). However the plants of Verbenaceae usually produce iridoid compounds and often the phenolic glycoside orobanchin but have a low occurrence of diterpenoids (Takhatajan, 1997). The isolation of the four diterpenoids suggests that genus *Callicarpa* (Subfam .Viticoideae Briq) should be separated from Verbenaceae or be placed in Labiatae. This is consistent with the taxonomic treatment proposed by Cantino (1992) which included the Labiatae sensu Briquet (1895–1897), plus subfamilies Caryopteridoideae, Chloanthoideae, Viticoideae and tribe Monochileae (subfamily Verbenoideae) of the Verbenaceae in a broadly circumscribed Labiatae s.l. This was then supported by Steven's *rbcL* sequence analyses (Steven and Richard, 1997) and Liang's evidences from floral organogenesis (Liang et al., 2001), respectively, as well.

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