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Chemical constituents from *Artemisia argyi* and their chemotaxonomic significance

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ABSTRACT

Twelve compounds, including one monoterpene (**1**), two sesquiterpene lactones (**2**–**3**), six flavonoids (**4**–**9**), one phenolic glycoside (**10**), one chromone (**11**) and one phenolic acid (**12**), were isolated and identified from the leaves of *Artemisia argyi*. Compounds **1**–**2**, **4** and **6**–**7** have not been recorded before in this plant. Compounds **3**, **9** and **11** were isolated from the genus *Artemisia* for the first time. This paper is the first report on the presence of compound **10** in species of Asteraceae. In addition, the chemotaxonomic significance of these compounds was summarized.

1. Subject and source

The genus *Artemisia*, one of the largest genera belonging to the family Asteraceae, consists of about 400 species. It is widely distributed in Europe, North America, Asia and South Africa (Lee et al., 2003; Mahmoud and Ahmed, 2006). There are 186 species along with 44 varieties grown in China, and they are nearly distributing all over the country (Lin, 1991). Some species of this genus such as *Artemisia argyi* Levl. et Vant. (Tan and Jia, 1992), *Artemisia annua* Linn. (Tu et al., 1982; Klayman, 1985; Baraldi et al., 2008) and *Artemisia capillaris* Thunb. (Wu et al., 1998, 2001) are used as famous traditional Chinese medicines for the treatment of malaria, hepatitis, cancer and infections by fungi, bacteria, and viruses (Kim et al., 2002).

A. argyi is a well-known medicinal herbaceous plant distributing throughout China (Zhou et al., 2000). The leaves of *A. argyi* were collected in May 2010 from Taiping town, Hubei province of China, and authenticated by Prof. Sui-Qing Chen (Department of Pharmacognosy, Henan College of Traditional Chinese Medicine). A voucher specimen (2010-Y0506) is deposited in the herbarium of Xinxiang Medical University, Xinxiang, China.

2. Previous work

Although phytochemical studies on *A. argyi* started in 1980s, most of the published data involved in the analysis of the chemical composition of essential oil of *A. argyi* from the different areas in China or discrimination it from related Asteraceae

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herbs (Hong et al., 1996; Yin et al., 1999; Guo, 2001; Jiang et al., 2009). Some other studies of the chemical constituents of *A. argyi* have led to the identification of sesquiterpenoids (Kim et al., 2002; Lee et al., 2002), flavonoids (Nakasugi et al., 2000; Ji et al., 2009, 2010), triterpenoids (Lao et al., 1984; Tan and Jia, 1992), sterols (Ji et al., 2010), and coumarin (Ji et al., 2010).

3. Present study

The air-dried and powdered leaves of *A. argyi* (7 kg) were extracted three times with 80% aqueous ethanol at room temperature. The combined extract was concentrated under reduced pressure to give the gross extract (920 g). Then, this residue was dissolved in water and partitioned sequentially with CH_2Cl_2, EtOAc and *n*-BuOH, respectively.

The CH_2Cl_2 extract (70 g) was subjected to column chromatography (CC) on silica gel and eluted with a gradient of petroleum ether–EtOAc (1:0, 20:1, 10:1, 5:1, 2:1, 1:1, 1:2 V/V) to give fractions D1–D7. Fraction D2 was chromatographed over silica gel CC and purified by CC on Sephadex LH-20 (MeOH) to afford compound **1** (10.6 mg). From fraction D4, compounds **2** (15.1 mg) and **3** (8.4 mg) were obtained through CC on silica gel and eluted with petroleum ether–EtOAc (20:1 and 10:1), respectively. Compounds **4** (25.2 mg), **5** (30.5 mg) and **6** (27.1 mg) were isolated from fraction D5 by repeated CC over silica gel and gradiently eluted with CH_2Cl_2–Acetone (50:1–10:1).

The EtOAc extract (168 g) was chromatographed on a silica gel column and eluted with petroleum ether–EtOAc (5:1–0:1) and EtOAc–MeOH (1:0–0:1) to afford 8 fractions (E1–E8). Fraction E2 was separated by CC on silica gel eluted with a gradient of CH_2Cl_2–Me_2CO (20:1–1:1) to afford compounds **7** (13.8 mg) and **11** (21.9 mg). Compounds **8** (11.4 mg) and **12** (20.6 mg) were fully purified from fraction E4 by repeated CC on silica gel and Sephadex LH-20 ($CHCl_3$–MeOH, 1:1). Fraction E7 was subjected to CC on Sephadex LH-20 eluted with MeOH–H_2O (10:1) to yield compounds **9** (6.9 mg) and **10** (10.1 mg).

The structures of the isolated compounds were elucidated on the basis of their spectroscopic data (IR, MS, 1D and 2D NMR), and by comparison of their spectroscopic data with those reported in the literature. Their structures were identified as 2α,5α-dihydroxy-β-pinene (**1**) (Huneck et al., 1986), 1β,2β-epoxy-3β,4α,10α-trihydroxyguaian-6α,12-olide (**2**) (Jakupovic et al., 1988; Zan et al., 2010), tanciloide (**3**) (Öksüz, 1990), casticine (**4**) (Han et al., 2007), eupatilin (**5**) (Deng et al., 2004a), 6-methoxytricin (**6**) (Martinez et al., 1987), 6,4′-dimethoxyl-scutellarin (**7**) (Lou et al., 2002), eriodictyol (**8**) (Liu et al., 2010), jaceosidin 7-β-glucoside (**9**) (Merfort, 1988), shimobashiraside C (**10**) (Murata et al., 2012), 5,7-dihydroxy-chromone (**11**) (Yang et al., 2008), and salicylic acid (**12**) (Xin et al., 2008) (Fig. 1). So far, only ^{1}H NMR data of compound **1** was reported. Therefore, its complete NMR data is displayed as follows.

Compound **1** was obtained as colourless amorphous solid. ESI-MS *m/z*: 169 $[M + H]^+$. ^{1}H NMR (400 MHz, in acetone-d_6/$CDCl_3$, 2:1) δ: 4.92 (1H, br s, H-7a), 4.68 (1H, br s, H-7b), 4.28 (1H, br s, H-5), 4.26 (1H, overlap, H-2), 4.04 (1H, br s, OH-5), 3.37 (1H, br s, OH-2), 2.43 (1H, br d, J = 6.4 Hz, H-6), 2.23 (1H, ddd, J = 2.3, 8.2, 14.4 Hz, H-3β), 1.89 (1H, m, H-4), 1.77 (1H, br d, J = 3.8, 14.4 Hz, H-3α), 1.48 (3H, s, H_3-10), 0.55 (3H, s, H_3-9). ^{13}C NMR (100 MHz, in acetone-d_6/$CDCl_3$, 2:1) δ: 155.9 (C-1), 111.4 (C-7), 74.4 (C-5), 66.5 (C-2), 57.7 (C-6), 46.6 (C-4), 39.9 (C-8), 36.2 (C-3), 28.1 (C-10), 23.9 (C-9).

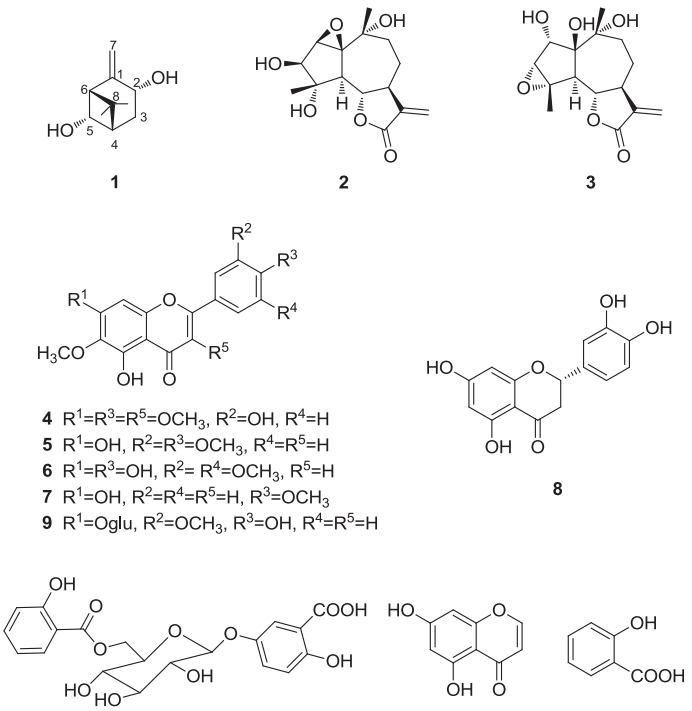

Fig. 1. Chemical structures of the isolated compounds from *Artemisia argyi*.