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Phenolic constituents from the leaves of *Syzygium forrestii* Merr. and Perry

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1. Subject and source

Syzygium forrestii Merr. and Perry, an evergreen broad-leaved tree distributed mainly in the southwest of China, was collected from the Puer City in Yunnan Province of China, in June 2009, and identified by Professor X. Chen, Kunming Institute of Botany, Chinese Academy of Sciences. A voucher specimen (No. KIB-ZL- 200902) of this collection was deposited in the State Key Laboratory of Phytochemistry and Plant Resources in West China, Kunming Institute of Botany, Chinese Academy of Sciences.

2. Previous work

The genus *Syzygium* (Myrtaceae) comprises about 500 species, mostly distributed in the tropic region of the world. Previous chemical investigation has demonstrated the presence of triterpenes (Djoukeng et al., 2005; Kuiate et al., 2007), hydrolysable tannins (Nonaka et al., 1992; Tanaka et al., 1996, 1993a), anthocyannins (Nonaka et al., 1992), flavonoids (Kuo et al., 2004; Resurreccion-Magno et al., 2005), chromone derivatives (Tanaka et al., 1993a; Toda et al., 2000), phenylpropanoids (Miyazawa and Hisama, 2003; Tanaka et al., 1993a), and phloroglucinols derivatives (Zou et al., 2006) in the genus *Syzygium*.

S. forrestii, distributed mainly on the mountain slopes with altitude range from 800 to 2400 m, is endemic to Yunnan Province, China. So far, there is no chemical investigation on this species.

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3. Present work

The air-dried leaves of *S. forrestii* (4.5 kg) were extracted with 80% aqueous acetone at room temperature (three times, each one a week). The crude extract was suspended in H_2O, and then partitioned with petroleum ether and EtOAc, successively. The EtOAc-soluble fraction (140 g) was subjected to Sephadex LH-20 column chromatography (CC), eluting with MeOH/H_2O (0:1–1:0) to give six fractions (fr. 1–6). Fr. 3 (10 g) was applied to MCI-gel CHP20P (MeOH/H_2O, 0:1–9:1), silica gel ($CHCl_3$/MeOH, 95:5–9:1), and Toyopearl HW-40F (MeOH/H_2O, 3:7–6:4) CC to yield **9** (4 mg), **14** (9 mg), **15** (16 mg), **17** (9 mg), and **18** (200 mg). A portion (10 g) of fr. 4 (47 g) was chromatographed over MCI-gel CHP20P (MeOH/H_2O, 4:6–6:4), silica gel

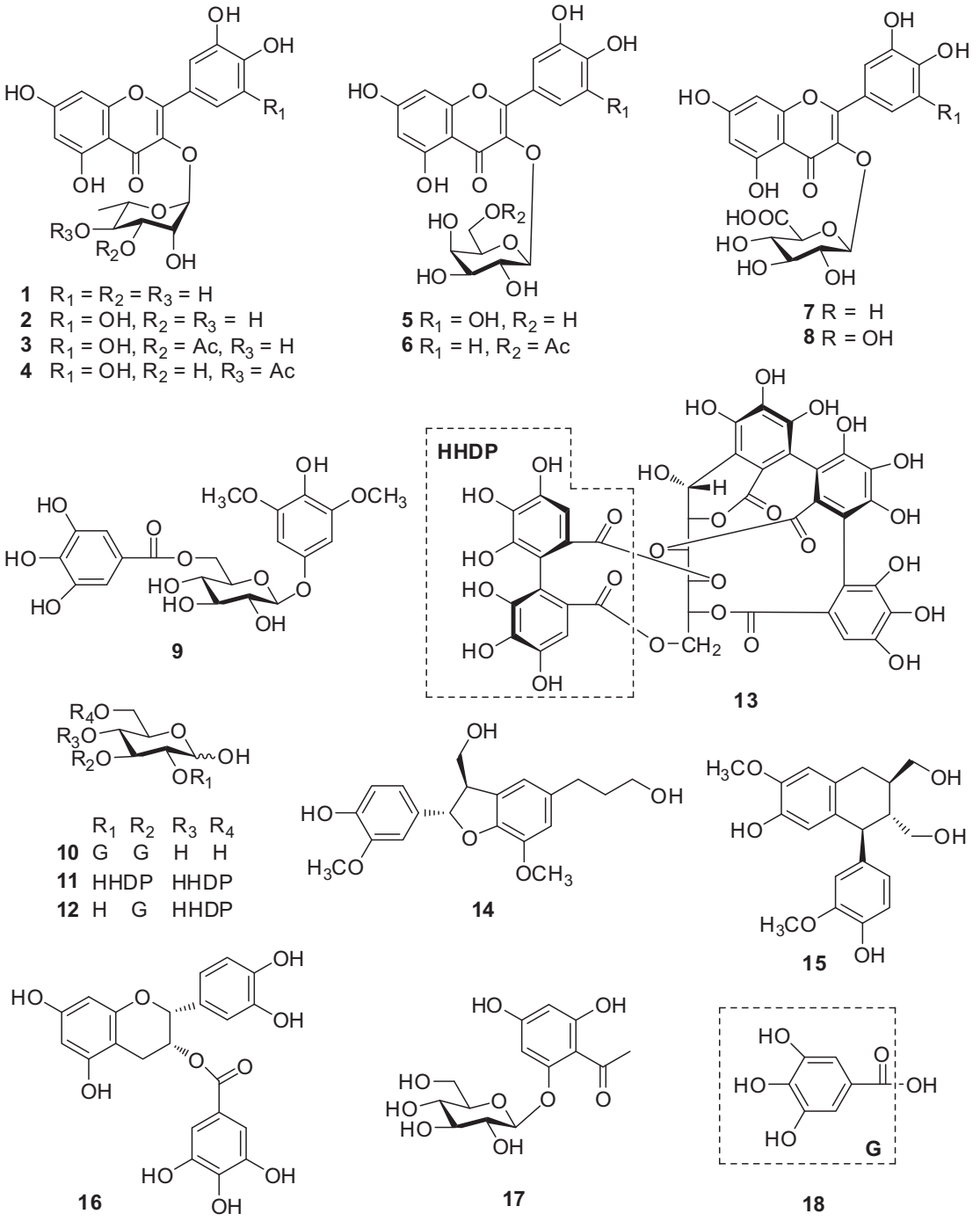

Fig. 1. Structures of compounds **(1-18)** isolated from the leaves of *S. forrestii*.

($CHCl_3$/MeOH, 95:5–9:1), and Rp-8 (MeOH/H_2O, 4:6–7:3) to give **1** (8 mg), **2** (80 mg), **3** and **4** (32 mg), **5** (20 mg), and **6** (14 mg). Repeated CC of fr. 5 (17 g) over MCI-gel CHP20P (MeOH/H_2O, 0:1–4:6) and Sephadex LH-20 (MeOH/H_2O, 4:6–1:0) afforded **11** (42 mg) and **16** (22 mg).

The water-soluble fraction (300 g) was chromatographed over Sephadex LH-20 (MeOH/H_2O, 0:1–1:0) to give six fractions (fr. A-F). Fr. B (28 g) was subjected to repeated CC over MCI-gel CHP20P (MeOH/H_2O, 0:1–3:7) to yield **8** (420 mg). Repeated CC over MCI-gel CHP20P (MeOH/H_2O, 0:1–3:7) and Sephadex LH-20 (MeOH/H_2O, 2:8–8:2) afforded **7** (30 mg) from Fr. C (31 g), **13** (52 mg) from Fr. D (7 g), and **10** (6 mg) and **12** (30 mg) from Fr. E (7 g), respectively.

The isolated compounds were identified as quercetin 3-*O*-α-L-rhamnopyranoside (**1**), myricitrin (**2**), myricetin 3-*O*-(3″-*O*-acetyl)-α-L-rhamnopyranoside (**3**) (Agnihotri et al., 2008), myricetin 3-*O*-(4″-*O*-acetyl)-α-L-rhamnopyranoside (**4**) (Timbola et al., 2002), myricetin 3-*O*-β-D-galactopyranoside (**5**), quercetin 3-*O*-(6″-*O*-acetyl)-β-D- galactopyranoside (**6**) (Chosson et al., 1998), quercetin 3-*O*-β-D-glucuronopyranoside (**7**), myricetin 3-*O*-β-D-glucuronopyranoside (**8**), 1-*O*-(3,5-dimethoxy-4-hydroxy- phenyl)-6-*O*-galloyl-β-D-glucuronopyranoside (**9**) (Ishimaru et al., 1987), nilocitin (**10**) (Nawwar et al., 1984), pedunculagin (**11**) (Tanaka et al., 1993b), gemin D (**12**), castalagin (**13**) (Nonaka et al., 1990), myrciaphenone A (**17**) (Chosson et al., 1998), and gallic acid (**18**), respectively, on the basis of their mass spectrometry, UV, ^1H and ^{13}C NMR spectroscopic analysis, together with the comparison with literature data (Fig. 1). Compounds **3** and **4** existed as a mixture, while (−)-dihydrodehydro- diconiferyl alcohol (**14**), (+)-isolariciresinol (**15**), (−)-epicatechol-3-*O*-gallate (**16**) (Nonaka et al., 1992) were also confirmed by their optical rotation values.

4. Chemotaxonomic significance

To the best our knowledge, it is the first report on the chemical constituents of *S. forrestii*. The isolated compounds from *S. forrestii* are assigned as flavonoids (**1**–**8**), hydrolysable tannins (**9**–**13**), lignans (**14**–**15**), and simple phenolic compounds (**16**–**18**). Among them, compounds **3**–**4**, **6**–**9**, **14**–**15** and **17** are new for the genus *Syzygium*, while compounds **3**, **6** and **9** are reported from the family Myrtaceae for the first time. The existence of acetylated glycosidic flavonoids (**3**, **4** and **6**) in genus *Syzygium*, after genus *Eugenia* and *Eucalyptus* (Myrtaceae), was not a coincidence.

The family Myrtaceae is characterized by tannins and flavonols as the major chemical constituents, of which the myricetin derivatives appeared quite frequently in this family (Amaral et al., 2001). The isolation of myricetin derivatives (**2**, **3**, **4**, **5**, and **8**) and ellagitannins (**11**, **12** and **13**) evidenced its taxonomic position under the family Myrteceae from the point view of chemotaxonomy. The main isolated flavonoid, myricitrin (**2**), has also been reported from *Syzygium levinei* and *Syzygium samarangense* (Kuo et al., 2004; Zou et al., 2006), while (−)-epicatechol-3-*O*-gallate (**17**) was found from *S. samarangens* (Nonaka et al., 1992). In addition, the hydrolysable tannins, nilocitin (**10**), pedunculagin (**11**), and gemin D (**12**), were reported from *Syzygium aromaticum* (Tanaka et al., 1993a).

The results indicate that *S. forrestii* has some similar flavonoids and hydrolysable tannins with other species of *Syzygium*. Flavonoid glycoside **2** (myricitrin) and hydrolysable tannins **10**–**12** can serve as the chemosystematic markers of the genus *Syzygium*.

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