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Iridoids from the flowers of *Gardenia jasminoides* Ellis and their chemotaxonomic significance

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ABSTRACT

Phytochemical investigation of the flowers of *Gardenia jasminoides* Ellis (Rubiaceae *Gardenia*) resulted in the isolation and identification of four iridoid aglycones (1–4) and eleven iridoid glycosides (5–15). This is the first report of the occurrence of these compounds in the genus *Gardenia*: garjasmine (1), dunnisin (2), α -gardiol (3), β -gardiol (4), diffusoside A (6), diffusoside B (7), genameside C (13), and deacetylasperulosidic acid (14). The chemotaxonomic significance and biosynthetic pathways of these iridoid aglycones (1–4) and iridoid glycosides (5–15) are summarized. Iridoid aglycones and iridoid glycosides are considered as important chemotaxonomic markers in Rubiaceae and this work indicates that the iridoid aglycones (1–4) have a limited distribution in the Rubiaceae.

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1. Subject and source

Gardenia jasminoides Ellis (Rubiaceae), is an evergreen shrub, grows in the south of the Yangtze River of China. The flowers of *G. jasminoides* are used for the remedy for the cough and epistaxis (Chinese Materia Medica Compilation Committee of State Administration of Traditional Chinese Medicine, 1999). The flowers were collected in Lishui County of Nanjing City of China and identified by Dr. Huan-yang Qi. A voucher specimen (No.2012G001) was deposited in the Key Laboratory for Natural Medicine of Gansu Province, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, China.

2. Previous work

The genus *Gardenia* includes about 250 species, of which 5 species are native to China (Flora Compilation Committee of Chinese Academy of Science, 2005). *G. jasminoides*, *G. jasminoides* var. *grandiflora* and *G. stenophylla* are used in the Chinese folk medicine (Chinese Materia Medica Compilation Committee of State Administration of Traditional Chinese Medicine, 1999). The fruits of *G. jasminoides*, named as “Zhi-Zi”, are used as a traditional Chinese medicine (Chinese Pharmacopeia Committee, 2010). Previous phytochemical investigations on *G. jasminoides* resulted in the isolation of iridoids (Inouye et al., 1970a,b, 1974; Zhao et al., 1994; Fu et al., 2008; Yu et al., 2009), flavonoids (Kim et al., 2006), diterpenoids (Choin

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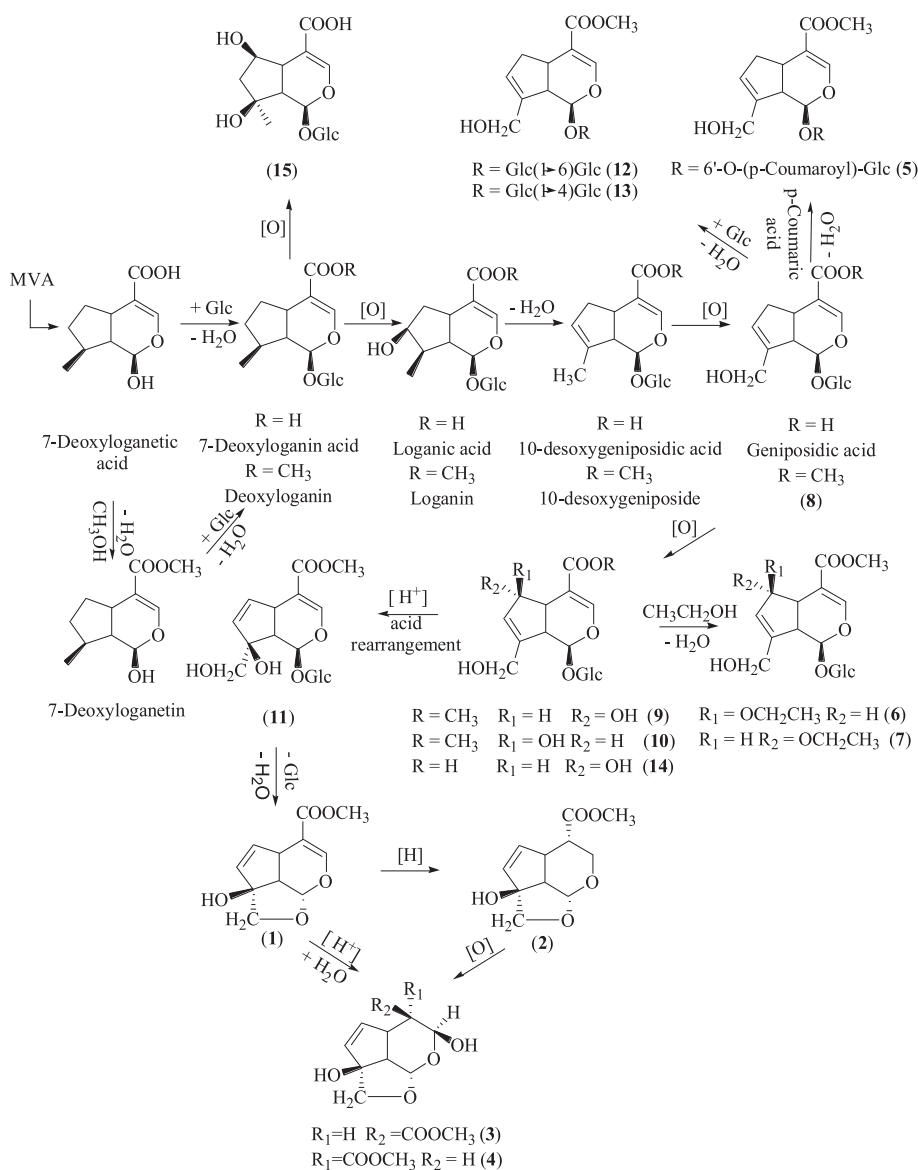


Fig. 1. Proposed biosynthetic pathway of iridoids in the flowers of *G. jasminoides*.

et al., 2001), and triterpenoids (Fu and Wang, 2011) from the fruits, and iridoids (Ragasa et al., 2007) and triterpenoids (Tai et al., 1989) from the flowers.

3. Present work

The air-dried flowers of *G. jasminoides* (6.0 kg) were extracted by 95% ethanol (v/v) and the filtrate was concentrated under reduced pressure. Then the extract (1.2 kg) was suspended in H₂O and extracted with petroleum ether and CHCl₃. The CHCl₃-soluble fraction (210.3 g) was subjected to silica gel column chromatography (CC) eluting with petroleum ether-EtOAc (v/v 20:1 → 0:1) to afford six fractions (I–VI). Fraction IV was purified on Sephadex LH-20 eluting with CHCl₃-MeOH (v/v 1:1), followed by silica gel CC (CHCl₃-acetone, v/v 10:1) and ODS CC (MeOH-H₂O, v/v 80:20) to yield **1** (15 mg) and **2** (10 mg). Fraction V was subjected to silica gel CC eluting with CHCl₃-acetone (v/v 7:1), followed by prep. HPLC (MeOH: H₂O, v/v 76:24) to afford **3** (20 mg) and **4** (16 mg). The remaining aqueous residue (705.6 g) was subjected to D101 macroporous resin CC, eluting with H₂O-EtOH. The H₂O-EtOH (v/v 10:90) fraction was subjected to silica gel CC eluting with CHCl₃-MeOH (v/v 10:1 → 0:1) to give seven fractions (I–VII). Fraction II was subjected to ODS CC eluting with MeOH-H₂O (v/v 60:40) to obtain **5** (8 mg). Fraction III was subjected to ODS CC eluting with MeOH-H₂O (v/v 55:45) to obtain **6** and **7** mixtures (21 mg) and **8** (32 mg). Fraction V was purified by prep-HPLC eluting with MeOH-H₂O (v/v 25:75) to obtain **9** (10 mg), **10** (20 mg) and **11**

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