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Chemical Constituents from *Erigeron bonariensis* L. and their Chemotaxonomic Importance

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Abstract: The study of the chemical constituents of the whole plant of *Erigeron bonariensis* (L.) has resulted in the isolation and characterization of a new and nine known compounds. The known compounds were identified as stigmasterol (**1**), freideline (**2**), 1,3-dihydroxy-3*R*,5*R*-dicaffeoyloxy cyclohexane carboxylic acid methyl ester (**3**), 1*R*,3*R*-dihydroxy-4*S*,5*R*-dicaffeoyloxycyclohexane carboxylic acid methyl ester (**4**), quercitrin (**5**), caffeic acid (**6**), 3-(3,4-dihydroxyphenyl)acrylic acid 1-(3,4-dihydroxyphenyl)-2-methoxycarbonylethyl ester (**8**), benzyl O-β-D-glucopyranoside (**9**), and 2-phenylethyl-β-D-glucopyranoside (**10**). The aromatic glycoside, erigoside G (**7**) is reported as new natural compound. The above compounds were individually identified by spectroscopic analyses and comparisons with reported data. The chemotaxonomic studies of isolated compounds have been discussed.
Keywords: *Erigeron bonariensis*; natural products; chemotaxonomic studies.

1.Plant Source

Erigeron bonariensis (L.) is locally called “gulava” or “mrich booti” and is traditionally used in urine problems. It is a common weed distributed from plains to ca. 1800 m height in North-West Frontier Province, Punjab and Balochistan in Pakistan [1]. The whole plants of *Erigeron bonariensis* (L.) (Asteraceae) were collected from Oghi, Mansehra, Pakistan, in November 2002, and authenticated by Mr. Jan Alam (Taxanomist) at the Botany Department, University of Karachi, Pakistan. A voucher specimen (KUH G. H. No. 68220) has been deposited at the herbarium of the above Department.

2. Previous Studies

Quercetin and quercitrin were identified from the ether and ethyl acetate soluble fraction of *E. bonariensis* [2].

3. Present Study

The air-dried whole plant (24 kg) were extracted with MeOH and then dried in vacuo. The crude extract was suspended in water, and successively extracted with *n*-hexane, $CHCl_3$, EtOAc, and *n*-BuOH in turn. The $CHCl_3$ extract (108 g) was subjected to silica gel column eluting with n-hexane/$CHCl_3$ yielded stigmasterol (**1**, 16.0 mg) [3], freideline (**2**, 45.0 mg) [4], 1,3-dihydroxy-3*R*,5*R*-dicaffeoyloxy cyclohexane carboxylic acid methyl ester (**3,** 8.0 mg) [4], 1*R*,3*R*-dihydroxy-4*S*,5*R*-dicaffeoyloxycyclohexane carboxylic acid methyl ester (**4**, 3.0 mg) [4], quercitrin (**5**, 10.0 mg) [5], caffeic acid (**6**, 15.0 mg) [6], erigoside G (**7**, 3.0 mg) [7], and 3-(3,4-Dihydroxyphenyl)acrylic acid 1-(3,4-dihydroxyphenyl)-2-methoxycarbonylethyl ester (**8**, 7.0 mg) [8], benzyl O-β-D-glucopyranoside (**9**, 8.0 mg) [9] and 2-phenylethyl-β-D-glucopyranoside (**10**, 16.0 mg) [10]. The isolated compounds were identified by comparison

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of their physical and spectroscopic data (^{1}H and ^{13}C NMR) with those reported in the literature (Figure 1). The new compound erigoside G (**7**) was obtained as brown gum. The UV spectrum exhibited absorption maxima at 263 nm. Analysis of the HREIMS gave a molecular ion at *m/z* 302.1112 $[M]^+$, corresponding to the molecular formula $C_{13}H_{18}O_8$, supported by the ^{1}H NMR, ^{13}C NMR and DEPT analysis. From the ^{1}H–^{1}H-COSY, two protons appeared at δ 6.91 (1H, dd, *J* = 7.5, 2.0 Hz, H-4) and δ 6.81 (1H, d, *J* = 7.5 Hz, H-5) were coupling to one another while one proton appeared at δ 7.20 (1H, d, *J* = 2.0 Hz, H-2), suggesting the presence of ABX spin system of three aromatic protons. Proton appeared at δ 7.20 (1H, d, *J* = 2.0 Hz, H-2) showed HMBC interaction with carbons appeared at δ 131.2, 65.3 and 123.8, while proton at δ 6.91 (1H, dd ,*J* = 7.5, 2.0 Hz, H-4) showed HMBC interaction with carbons appeared at δ 149.9 and 131.2, suggesting the structure consist on 3,4-dihydroxybenzyl alcohol system. A signal at δ 4.77 (1H, d, *J* = 7.5 Hz, H-1′) was assigned to the anomeric proton of glucopyranoside moiety. The anomeric configuration was assigned to be β on the basis of large coupling constant. In ^{13}C-NMR spectrum, the signals for β-D-glucopyranoside moiety were observed at δ 104.5, 75.0, 77.7, 71.4, 78.4, and 62.6. The HMBC interaction of anomeric proton δ 4.77 (1H, d, *J* = 7.5 Hz, H-1′) with C-1 confirmed the structure of compound **7** as 3,4-dihydroxybenzyl alcohol-3-*O*-β-D-glucopyranoside (erigoside G) (Figure 1). The aromatic glycoside (**7**) is new as a natural product but has been reported as a bio transformed product of 3,4-dihdroxybenzaldehyde by hairy root culture of *Pharbitis nil* [7].

Erigoside G (**7**): Brown gum. UV (CH_2Cl_2) λ_{max}, nm (log ε): 263 (2.10). IR (KBr) ν_{max}: 3200, 2963, 1606, 1465, 1061 cm^{-1}. ^{1}H NMR (600 MHz, CD_3OD): δ 3.40 (1H, t, *J* = 9.5 Hz, H-4′), 3.41 (1H, m, H-5′), 3.46 (1H, t, *J* = 9.0 Hz, H-3′), 3.49 (1H, t, *J* = 9.0 Hz, H-2′), 3.72 (1H, dd, *J* = 12.5, 5.0 Hz, H-6′), 3.90 (1H, dd, *J* = 12.5, 2.0 Hz, H-6′), 4.48 (2H, s, H-7), 4.77 (1H, d, *J* = 7.5 Hz, H-1′), 6.81 (1H, d, *J* = 7.5 Hz, H-5), 6.91 (1H, dd, *J* = 7.5, 2.0 Hz, H-4), 7.20 (1H, d, *J* = 2.0 Hz, H-2). ^{13}C NMR (CD_3OD) δ: 62.6 (C-6′), 65.1 (C-7), 71.4 (C-4′), 75.0 (C-2′), 77.7 (C-3′), 78.4 (C-5′), 104.5 (C-1′), 117.0 (C-2), 118.2 (C-5), 123.8 (C-4), 134.7 (C-3), 146.8 (C-6), 147.9 (C-1). HREIMS: *m/z* 302.1112 $[M]^+$ (Calcd. 302.1002 for $C_{13}H_{18}O_8$).

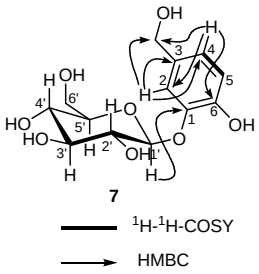

Figure 1. Selected ^{1}H-^{1}H COSY and HMBC correlations of compound **7.**

4. Chemotaxonomic significance

Erigeron is a genus of about 390 species of flowering plants in the family Asteraceae. It represents one of the foremost examples of intercontinental plant invasions that have resulted in a number of taxonomic problems, especially in distinguishing it from *Conyza* [11]. We have previously investigated the title species and considered to be a synonym of *Conyza bonariensis* (L.) [12]. But actually *Erigeron* and *Conyza* are two different genera [11]. From previous [12] as well as present investigation, on the basis of chemotaxonomy we have concluded that the title species is *Erigeron bonariensis.* Because we consider that the phenolic constituents and caffeoyl derivatives may indicate it has a closer relationship to the genus *Erigeron* than the other species of *Conyza*. The flavonoids, caffeoyl derivatives and triterpenes identified here are in agreement with the chemical profile of the genus *Erigeron* (Table 1). Compounds **1-6** identified have already been isolated from *Erigeron* species as shown in previous studies, specially frideline (**2**) and their derivatives which seem to be characteristic secondary metabolites of the genus *Erigeron* (see Table 1).

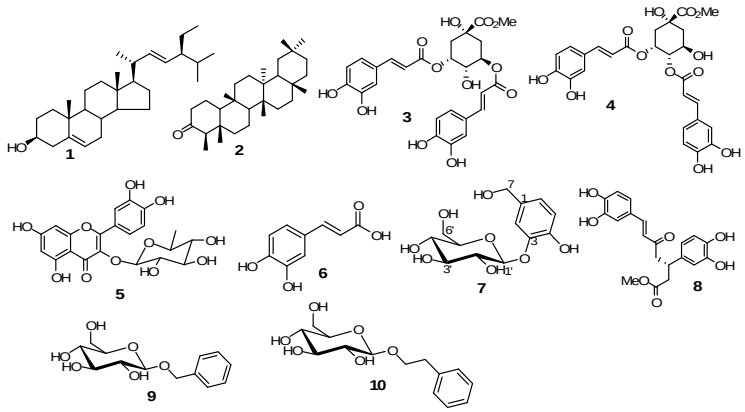

Figure 2. Structure of compounds **1-10** isolated from *Erigeron bonariensis* L.

Caffeoylquinic acids are widespread among Asteraceae, and there methyl esters **3** and **4** have already been isolated from the *E. breviscapus* [13]. This is the first report of compounds **8-10** in plants of the genus *Erigeron*. This might well be explained since most of the previous studies focused on medium polarity extracts of *Erigeron*. Isolation of compound **10** was also reported from *Mikania hirsutissima* and *Gymnaster koraiensis* of Asteraceae [14,15]. Similarly isolation of compound **9** was reported from *Helichrysum conglobatum* and *Baccharis dracunculifolia* of the same family [16,17] which is particularly interesting since this strengthens the chemotaxonomic relationship of genus *Erigeron* and *Helichrysum* and *Baccharis*. Compound **8** is an uncommon compound and was previously isolated only from *Isodon excisus* [8] but it is a caffeoylic conjugate proving the agreement of given subject with Asteraceae. Also presences of quercitrin (**5**) [5], and absence of pyromeconic acid and its derivatives provides the chemotaxonomic difference among *E. bonariensis* L. and other *Erigeron* species [2]. *E. bonariensis* L. represents one of the foremost examples of intercontinental plant invasions that have resulted in a number of taxonomic problems, especially in distinguishing it from *Conyza* Less. (Asteraceae) [11]. Previous phytochemical studies on *Conyza* species have led to the isolation of about flavonoids mainly including rutin, quercetin-3-O-glucoside, and quercetin [18]. In contrast, the plants belonging to *Erigeron* are described as containing flavonoids (scutellarin as the major components) (Table 1) [19-21], caffeoyl derivatives (Table 1), which are regarded as the characteristic constituents of the genus. Finally, it should be noted that this chemotaxonomic study (including Table 1) could be helpful for a better understanding in regard to the controversial taxonomy of the genus *Erigeron*, whose taxonomic classification have been altered by the establishment of synonymies and the transference of plants from and to the genus [22].

Table 1. Flavonoids, triterpenes, caffeoylic derivatives, and steroids isolated from genus *Erigeron*

Flavones	
Kaempferol	*E. multiradiatus* [23]
	E. multiradiatus [24]
5,7,4'-Trihydroxyflavanone	*E. breviscapus* [25]
Quercetin	*E. acer* [26], *E. bonariensis* [5]
7-Hydroxy-4',5-dimethoxyflavone,	*E. annuus* [27]
Quercitrin	*E. bonariensis* [5]
3,5,6,4'- Tetrahydroxy-7-methoxyflavone	*E. breviscapus* [25]
Apigenin	*E. breviscapus* [25]
3,5,6,7,4'-Pentahydroxyflavone	*E. breviscapus* [25]
Apigenin-7-O-β-D-glucuronide butyl ester	*E. multiradiatus*. [24]
Kaempferol-7-O-α-L-rhamnopyranoside	*E. multiradiatus* [24]
Kaempferol-3-O- β-D-glucopyranoside-7-O- α -L-rhamnopyranoside	*E. multiradiatus* [24]
Apigenin-7-O-β-D-glucoside	*E. multiradiatus* [28]
Apigenin-7-glucuronide	*E. annuus* [27]
4',5-dimethoxyapigenin,	*E. annuus* [27]
5,4'-Dihydroxyflavone-7-O-β-D-pyranglycuronate butyl ester	*E. breviscapus* [29]
Scutellarin	*E. breviscapus* [30]
Apigenin 7-O-β-D-pyranoglucuronide	*E. breviscapus* [31]
Plantaginin	*E. multiradiatus* [28]
Scutellarein-7-O-β-D-glucoside	*E. multiradiatus* [28]
Wogonin	*E. multiradiatus* [23]
Luteolin	*E. multiradiatus* [28]
Triterpenes	
β-Amyrin	*E. acer* [26], *E . sumatrensis* [32]
α-Amyrin	*E. acer* [26], *E. sumatrensis* [32]
3β,23,28-Trihydroxy-12-oleanene acetonide	*E. annuus* [33]
Taraxerol	*E. acer* [26], *E. sumatrensis* [32]
Lupeol	*E. acer* [26], *E. sumatrensis* [32]
Friedelin	*E. acer* [26], *E. sumatrensis* [32]
Epifriedelinol	*E. acer* [26], *E. sumatrensis* [32]
Fernenol	*E. acer* [26], *E. sumatrensis* [32]
24-Methylenelanost-8-en-3-β-ol	*E. acer* [26], *E. sumatrensis* [32]

Glutinol	*E. acer* [26], *E. sumatrensis*[32]
Schottenol	*E E. acer* [26], *E. sumatrensis*[32]
Simiarenol	*E. acer* [26], *E. sumatrensis*[32]
Steroids	
Ergosterol peroxide	*E. annuus L* [35], *E. multiradiatus* [24]
Stigmasterol	*E. annuus* [33], *E. acer* [26], *E. sumatrensis* [32]
Stigmast-5-ene-3β,7α-ol	*E. annuus* [33]
Stigmast-4-ene-3β,6α -ol	*E. annuus* [33]
Stigmast-7,24-dien-3β-ol	*E. annuus* [33]
Stigmasterol glucoside	*E. multiradiatus* [24]
Stigmast-7-en-3β -ol	*E. Canadensis* [36]
Stigmasta-7,22-dien-3-one	*E. Canadensis* [36]
Stigmast-7-en-3-one	*E. Canadensis* [36]
α-Spinasterol	*E. Canadensis* [36]
Caffeoyl derivatives	
Caffeic acid	*E. breviscapus* [37], *E. acer* [26]
1,5-Dicarffeoylquinic ester	*E. breviscapus* [13]
3,5-Dicarffeoylquinic ester	*E. breviscapus* [13]
4,5-Dicarffeoylquinic ester	*E. breviscapus* [13]
Erigoster A	*E. breviscapus* [13]
Erigoster B	*E. breviscapus* [13]
Erigeroside	*E. breviscapus* [37]
1-O-(4-Pyranon-3-yl)-6-O-caffeoyl-α-D-pyranoglucose	*E. breviscapus* [38]
1*R*,3*R*-Dihydroxy-4*S*,5*R*-dicaffeoyloxy cyclohexane carboxylic acid methyl ester	
Erigeside I	*E. breviscapus* [39]
Erigeside II	*E. breviscapus* [39]
Methylcaffeate	*E. breviscapus* [40]
6'-O-Caffeylerigeroside	*E. multiradiatus* [28]
(1*R*,3*R*)-Dihydroxy-(4*S*,5*R*)-dicaffeoyloxycyclohexane carboxylic acid methyl ester	*E. breviscapus* [41]
1,4-Dihydroxy-(3*R*,5*R*)-dicaffeoyloxycyclohexane carboxylic acid methyl ester	*E. breviscapus* [41]
Ethylcaffeate	*E. breviscapus* [41]
Miscellaneous aromatic compounds	
p-Hydroxybenzoic acid	*E. multiradiatus* [23], *E. breviscapus* [30], *E. multiradiatus* [28]
1-Hydroxy-2,3,5-trimethoxyxanthone	*E. multiradiatus* [23]
3,5-Dimethoxy-3',4':2",3"-dihydro-furanchalcone	*E. multiradiatus* [23]
3,4-Dihydroxybenzoic acid	*E. breviscapus* [40]
β-(p-Methoxyphenyl)acrylic acid	*E. breviscapus* [40]
3,5-Dimethoxy-4-hydroxy benzene carbonic acid	*E. breviscapus* [42]
6-Methoxy coumarin-7-O-β-D-pyranglycoside	*E. breviscapus* [42]
Vanillic acid	*E. multiradiatus* [28]
Scopoletin	*E. breviscapus* [41]
Isoscopoletin	*E. breviscapus* [41]
3,5-Dimethoxy benzene carbonic acid-4-O- β-D-pyranglucose	*E. breviscapus* [29]
Emodin	*E. multiradiatus* [23]
Pyromeconic acid	*E. breviscapus* [41]

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