Iridoids from Scrophularia Genus

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Iridoid Glycosides, Scrophularia, Scrophularia scorodonia

We report here an updated summary about iridoid composition of a series from the genus Scrophularia which have been investigated until now from a phytochemistry point of view. In addition a list is included about iridoids isolated in our laboratory from different plant parts of Scrophularia scorodonia L., which are compared with iridoids from some species of the Scrophularia genus. The present study may serve as a current information to researchers working on phytochemistry and pharmacological aspects from the Scrophularia genus and possibly to serve as a new starting point for future investigations.

Iridoid glycosides such as harpagoside, aucuboside and harpagide are of particular interest to botanists because of their potential value in chemosystematics to elucidate phylogenetic relationships (Bianco, 1990; Luczak and Swiatek, 1978).

The Scrophularia genus is represented by more than 300 species. But only a few species have been investigated from a phytochemical and pharmaceutical point of view. We have now examined species from genus Scrophularia which have been investigated from a phytochemical point of view, and report here the results of the literature search. In genus Scrophularia the following iridoid glycosides have been found till now: Aucuboside, 6'-0-ß-glucosylaucuboside and scropheanoside I in: Scrophularia: Scrophularia scorodonia (Berdini et al., 1991), Scrophularia alata Gilib (Swiatek and Broda, 1967), Scrophularia grossheimi (Akhmedov, 1970), Scrophularia nodosa (Weinges and von der Eltz, 1978), Scrophularia scorodonia (Qian et al., 1992; Ming et al., 1999), Scrophularia leucocolada (Saatov and Abdullaev, 1996), Scrophularia vernalis (Swiatek and Krzaczek, 1976) and Scrophularia koraiensis nakai (Pachaly et al., 1994) (Table II). Acylated 6-O-α-L-rhamnopyranosylcatalpol are common in genus Scrophularia: Scrophularia scopoli var. scopoli (Calis et al., 1988), Scrophularia nodosa (Weinges and von der Eltz, 1978), Scrophularia spicata (Zhang et al., 1992), Scrophularia ilwensis (Calis et al., 1993), Scrophularia auriculata (Giner et al., 1991; 1998), Scrophularia koelzii (Bhandari et al., 1992) and Scrophularia koraiensis nakai (Pachaly et al., 1994) (Table III). Catalpol, methylcatalpol, jugol and laterioside were found only in some species : Scrophularia ningpoensis (Qian et al., 1992), Scrophularia vernalis (Swiatek and Krzaczek, 1976), Scrophularia lateriflora (Swiatek et al., 1981), Scrophularia ilwensis (Calis et al., 1993).

The presence of picroside I was detected only in one species, Scrophularia lateriflora (Swiatek et al., 1981) (Tables IV, V).

In the course of a search for biologically active substances in S. scorodonia we have isolated several iridoids from leaves and stems, flowers and roots from this species.

For leaves and stems we have reported, bartsioid, aucuboside, harpagoside, 8-O-cis-cinnamoyl-harpagide, 8-O-acetyl-harpagide and one new iridoid 6-O-α-L-(3'-O-acetyl-2'-O-trans-cinnamoyl)-...
Different catalpol derivates have been isolated in this species: 6-O-α-L-(3"-O-trans and cis p-coumaroyl)-rhamnopyranosylcatalpol and 6-O-α-L-(2"-O-trans-p-coumaroyl)-rhamnopyranosylcatalpol (saccatoside) have been reported in flowers; 6-O-α-L-(2"-O-acetyl-3",4"-O-di-trans-cinnamoyl)-rhamnopyranosylcatalpol (scropolioside B) and 6-O-methylcatalpol in roots from *S. scorodonia* (De Santos *et al.*, 1998).

It is of interest to compare the presence and distribution of some of these iridoids in different plant parts, because a different composition has been found in leaves, stems, flowers and roots (Table VI). We have detected the presence of harpagide for the first time in leaves, stems and roots from *S. scorodonia*. In roots, we have isolated and identified different iridoids: bartsioside, aucuboside, harpagoside and 8-O-acetyl-harpagide. These compounds have also been isolated from flowers of this species, except aucuboside. Scorodioside, the new iridoid glycoside isolated from leaves and stems, has also been identified in flowers. As shown in Table VI there is a different distribution between all plant parts.

Identification and assignment of the isolated compounds were performed on the basis of UV spectral data, 1H and 13CNMR.

**Results and Discussion**

We have compiled a first listing of iridoids occurrence in Scrophularia genus (Table VII). From the distribution of iridoids listed it can be concluded that the presence of aucuboside (52.9%), harpagide (47.05%), harpagoside (47.05%) and 8-O-acetyl-harpagide (41.17%) is not rare in species of the genus, and we have also isolated these compounds from *Scrophularia scorodonia*.

Table I. Aucuboside, scrophularioside, bartsioside and 10-O-β-glucosylaucuboside.

<table>
<thead>
<tr>
<th>Compound</th>
<th>R</th>
<th>Species</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aucuboside</td>
<td>R1=R2=R3=H</td>
<td><em>S. nodosa</em></td>
<td>(Weinges and von der Eltz, 1978)</td>
</tr>
<tr>
<td></td>
<td>R4=OH</td>
<td><em>S. grossheimi</em></td>
<td>(Akmedov, 1970)</td>
</tr>
<tr>
<td></td>
<td></td>
<td><em>S. lateriflora</em></td>
<td>(Swiatek <em>et al.</em>, 1981)</td>
</tr>
<tr>
<td></td>
<td></td>
<td><em>S. canina</em></td>
<td>(Berdini <em>et al.</em>, 1991)</td>
</tr>
<tr>
<td></td>
<td></td>
<td><em>S. ningpoensis</em></td>
<td>(Quian <em>et al.</em>, 1992)</td>
</tr>
<tr>
<td></td>
<td></td>
<td><em>S. ilwensis</em></td>
<td>(Calis <em>et al.</em>, 1995)</td>
</tr>
<tr>
<td></td>
<td></td>
<td><em>S. scorodonia</em></td>
<td>(Fernandez <em>et al.</em>, 1995 a)</td>
</tr>
<tr>
<td></td>
<td></td>
<td><em>S. alata</em></td>
<td>(Swiatek and Broda, 1967)</td>
</tr>
<tr>
<td></td>
<td></td>
<td><em>S. vernalis</em></td>
<td>(Swiatek and Krzuczek, 1976)</td>
</tr>
<tr>
<td>Scrophularioside</td>
<td>R1=R2=H</td>
<td><em>S. lateriflora</em></td>
<td>(Swiatek <em>et al.</em>, 1981)</td>
</tr>
<tr>
<td></td>
<td>R3=C6H2CH=CH-CO</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>R4=OH</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10-O-β-glucosylaucuboside</td>
<td>R1=R3=H</td>
<td><em>S. canina</em></td>
<td>(Berdini <em>et al.</em>, 1991)</td>
</tr>
<tr>
<td></td>
<td>R2=β-D-GLUCOSA</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>R4=OH</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bartsioside</td>
<td>R1=R2=R3=R4=H</td>
<td><em>S. scorodonia</em></td>
<td>(Fernandez <em>et al.</em>, 1995 a)</td>
</tr>
<tr>
<td>Scropheanoside I</td>
<td>R1=R2=R3=H</td>
<td><em>S. koraiensis</em></td>
<td>(Pachaly <em>et al.</em>, 1994)</td>
</tr>
<tr>
<td></td>
<td>R4=4&quot;-O-(p-OCH3-)m-OH-C6H5-CH=CH-CO</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>β-D-RHAMNOSA</td>
</tr>
</tbody>
</table>
On the other hand, iridoids derived from aucuboside are rarer in the genus Scrophularia, they appear only in some species, such as scrophularioide in *S. lateriflora*, 10-O-ß-glucosylaucuboside in *S. canina* and scrophenoside I in *S. koraiensis* (Table I).

Moreover it is well known that species of Scrophularia contain catalpol derivates (Table III), but some of this iridoids have been reported only in one species: koelzioside in *S. koelzii*, scropolioside A and B in *S. scopolii* and *S. auriculata*, scrospioide B in *S. scopolii*, scrophenoside II and 6-O-ß-1-(2"-O-acetyl-3"-4"-O-di-p-methoxycinamoyl) rhamnopyranosylcatalpol in *S. koraiensis* and *S.auriculata*, respectively. Picroside I has been detected only in *S. lateriflora* and 6-O-(ß-D-xylo-
Table III. Acylated derivates of 6-O-α-L-rhamnopyranosylcatalpol.

<table>
<thead>
<tr>
<th>Compound</th>
<th>R</th>
<th>Species</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>6-α-L-rhamnopyranosyl-catalpol</td>
<td>R1=R2=R3=H</td>
<td>S. nodosa</td>
<td>(Weinges and von der Eltz, 1978)</td>
</tr>
<tr>
<td>6-0-α-L-(4'-O-acetyl-2', 3'-di-O-cinnamoyl)-rhamnopyranosyl-catalpol</td>
<td>R1=R2=C6H5-CH=CH-CO-</td>
<td>S. koeltzi</td>
<td>(Bhandari et al., 1992)</td>
</tr>
<tr>
<td>(koelzioside)</td>
<td>R3=CH3-CO-</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6-0-α-L-(2', 4'-di-O-acetyl-3'-O-p-methoxy-trans-cinnamoyl) -rhamnopyranosyl-catalpol</td>
<td>R1=R3=CH3-CO-</td>
<td>S. scopolii</td>
<td>(Calis et al., 1988)</td>
</tr>
<tr>
<td>(scropolioside A)</td>
<td>R2=CH3O-C6H5-CH=CH-CO-</td>
<td>var scopolii</td>
<td>(Giner et al., 1988)</td>
</tr>
<tr>
<td>6-0-α-L-(2'-O-acetyl-3', 4'-O-di-trans-cinnamoyl)-rhamnopyranosyl-catalpol</td>
<td>R1=CH3-CO-</td>
<td>S. scopolii</td>
<td>(Calis et al., 1998)</td>
</tr>
<tr>
<td>(scropolioside B)</td>
<td>R2=R3=C6H5-CH=CH-CO-</td>
<td>var. Scopolii</td>
<td>(De Santos et al., 1998)</td>
</tr>
<tr>
<td>6-0-α-L-(2', 4'-di-O-acetyl-3'-O-trans-cinnamoyl)-rhamnopyranosyl-catalpol</td>
<td>R1=R3=CH3-CO-</td>
<td>S. ilwensis</td>
<td>(Calis et al., 1993)</td>
</tr>
<tr>
<td>(scropolioside D)</td>
<td>R2=C6H5-CH=CH-CO-</td>
<td>S. spicata</td>
<td>(Zhang et al., 1992)</td>
</tr>
<tr>
<td>6-0-α-L-(2'-O-acetyl-3'-O-trans-cinnamoyl)-rhamnopyranosyl-catalpol</td>
<td>R1=CH3-CO-</td>
<td>S. koraensis</td>
<td>(Pachaly et al., 1994)</td>
</tr>
<tr>
<td>(scropolioside B)</td>
<td>R2=C6H5-CH=CH-CO-</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Scropheanoside II</td>
<td>R1=R2=R3=H</td>
<td>S. koraensis</td>
<td>(Pachaly et al., 1994)</td>
</tr>
<tr>
<td>6-0-α-L-(2'-O-acetyl-3', 4'-O-di-p-methoxycinnamoyl-</td>
<td>R1=CH3-CO-</td>
<td>S. auriculata</td>
<td>(Giner et al., 1991)</td>
</tr>
<tr>
<td>rhamnopyranosyl-catalpol</td>
<td>R2=R3=p-OCH3-m-OH-C6H5-CH=CH-CO-</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6-0-α-L-(3'-O-acetyl-2'-trans-cinnamoyl)-rhamnopyranosyl-catalpol</td>
<td>R1=C6H5-CH=CH-CO-</td>
<td>S. Scorodonia</td>
<td>(Fernandez et al., 1995 b)</td>
</tr>
<tr>
<td>(scrodesioside)</td>
<td>R2=R3=H</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6-0-α-L-(2'-O-trans-p-coumaroyl)-rhamnopyranosyl-catalpol</td>
<td>R1=H</td>
<td>S. scorodonia</td>
<td>(De Santos et al., 1998)</td>
</tr>
<tr>
<td>(saccatoside)</td>
<td>R2=R3=CH3-CO-</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6-0-α-L-(3'-O-trans and cis-p-coumaroyl)-rhamnopyranosyl-catalpol</td>
<td>R1=R3=H</td>
<td>S. scorodonia</td>
<td>(De Santos et al., 1998)</td>
</tr>
<tr>
<td>(saccatoside)</td>
<td>R2=HO-C6H5-CH=CH-CO-</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6-0-α-L-(2', 3'-di-O-acetyl-4'-O-p-methoxycinnamoyl)-rhamnopyranosyl-catalpol</td>
<td>R1=R3=CH3-CO-</td>
<td>S. auriculata</td>
<td>(Giner et al., 1988)</td>
</tr>
<tr>
<td>(scrovalentinoside)</td>
<td>R2=CH3O-C6H5-CH=CH-CO-</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

pyranosyl) methylcatalpol is unique in *S. ilwensis* (Table IV).

In *S. scorodonia* we have isolated three 6-O-α-L-rhamnopyranosylcatalpol derivates as saccatoside, and 6-O-α-L-(3'-O-trans and *cis*-*p*-coumaroyl) rhamnopyranosylcatalpol which have not been reported previously in this genus. Also, bartsioside and 8-O-*cis*-cinnamoyl-harpagide are not present.
Table IV. Catalpol, methylcatalpol and picroside I.

<table>
<thead>
<tr>
<th>Compound</th>
<th>R</th>
<th>Species</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Catalpol</td>
<td>R1=R2=R3=R4=H</td>
<td>S. lateriflora</td>
<td>(Swiatek et al., 1981)</td>
</tr>
<tr>
<td>Picroside I</td>
<td>R1=R3=R4=H</td>
<td>S. lateriflora</td>
<td>(Swiatek et al., 1981)</td>
</tr>
<tr>
<td></td>
<td>R2=C₆H₅CH=CH-CO-</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6-Methylcatalpol</td>
<td>R1=R2=R4=H</td>
<td>S. vernalis</td>
<td>(Swiatek and Krzaczek, 1976)</td>
</tr>
<tr>
<td></td>
<td>R3=CH₃-</td>
<td>S. lateriflora</td>
<td>(Swiatek et al., 1981)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>S. ningpoensis</td>
<td>(Quian et al., 1981)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>S. scorodonia</td>
<td>(De Santos et al., 1998)</td>
</tr>
<tr>
<td>6-O-((β-D-xylopiranosyl)-methylcatalpol</td>
<td>R1=R2=H</td>
<td>S. ilwensis</td>
<td>(Calis et al., 1993)</td>
</tr>
<tr>
<td></td>
<td>R3=CH₃-</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>R4=O-β-xylopiranosyl</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table V. Ajugol and laterioside.

<table>
<thead>
<tr>
<th>Compound</th>
<th>R</th>
<th>Species</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ajugol</td>
<td>R=H</td>
<td>S. lateriflora</td>
<td>(Swiatek and Broda, 1967)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>S. ilwensis</td>
<td>(Weinges and von der Eltz, 1978)</td>
</tr>
<tr>
<td>Laterioside</td>
<td>R=C₆H₅CH=CH-CO-</td>
<td>S. lateriflora</td>
<td>(Swiatek and Broda, 1967)</td>
</tr>
</tbody>
</table>

In Scrophularia genus, but we have isolated these compounds in *S. scorodonia*.

Of special interest is scorodioside, a new catalpol analog until now not reported as present in nature and which we have isolated from *S. scorodonia*.

Remarkably, catalpol, one of the most common iridoids, has not been found in the present investigation. This compound has been isolated in the genus Scrophularia only in *S. lateriflora* (Table IV).

With regard to the widespread occurrence of iridoids in members of genus Scrophularia, *Scrophularia scorodonia* fits well into the genus, but the iridoid patterns were some different between this species and others species from this genus. As shown in Table VII *S. scorodonia* is the species which contains the major variety of iridoids, with 11 iridoids among 31 found until now in this genus and with an unequal distribution in different plant parts as shown in Table VI.
Table VI. Iridoids from *Scrophularia scorodonia*.

<table>
<thead>
<tr>
<th>Iridoids</th>
<th>Scrophularia scorodonia</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Flowers</td>
</tr>
<tr>
<td>Aucuboside</td>
<td></td>
</tr>
<tr>
<td>Bartsioside</td>
<td>*</td>
</tr>
<tr>
<td>Harpagide</td>
<td></td>
</tr>
<tr>
<td>8-O-acetyl-harpagide</td>
<td>*</td>
</tr>
<tr>
<td>Harpagoside</td>
<td></td>
</tr>
<tr>
<td>8-O-cis-cinnamoyl-harpagide</td>
<td>*</td>
</tr>
<tr>
<td>Scropolioside B</td>
<td></td>
</tr>
<tr>
<td>Scorodioside</td>
<td></td>
</tr>
<tr>
<td>Saccatoside</td>
<td></td>
</tr>
<tr>
<td>6-O-α-L-(3’0-trans and cis-p-coumaroyl)-rhamnopyranosyl-catalpol</td>
<td></td>
</tr>
<tr>
<td>6-Methylcatalpol</td>
<td></td>
</tr>
</tbody>
</table>

So, bartsioside, harpagide, 8-O-acetyl-harpagide and harpagoside, are present in all plant parts. Catalpol derivatives are predominant in roots and flowers. Although scorodioside, the new iridoid isolated from *S. scorodonia*, is present in leaves, stems and flowers, it has not been detected in roots. Aucuboside, a common iridoid, was not found in flowers from this species, but it is present in leaves, stems and roots.

Iridoids are present in a number of folk medicines used as bitter tonics, sedatives, antipyretics, cough medicines, remedies for wounds, skin disorders and as hypotensives. Recently, intensive investigations on their bioactivity in general, and on their potential pharmacological activity in particular, have revealed that these compounds exhibit a wide range of bioactivity: cardiovascular, antihepatotoxic, choleretic, hypoglycemic and hypolipidemic, antiinflammatory, antispasmodic, antitumor, antiviral, immunomodulator and purgative activities (Ghisalberti, 1998). In addition, aucuboside is reported to exhibit antihepatotoxic, antispasmodic and antitumoral activities (Ghisalberti, 1998). The presence of 8-O-acetyl-harpagide may also be important, as the compound has shown antispasmodic and vasoconstrictor activities (Breschi et al., 1992). Catalpol derivatives have shown to possess a lot of pharmacological properties, these type of compounds also exhibit antiinflammatory activity. Studies on antiinflammatory activity of these catalpol derivatives and other iridoids isolated from *S. scorodonia* have exhibited an interesting activity (Bermejo et al., 1999).

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Table VII. Iridoids of the Scrophularia genus.

|   | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 21 | 22 | 23 | 24 | 25 | 26 | 27 | 28 | 29 | 30 | 31 |
|---|---|---|---|---|---|---|---|---|---|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
| **S. nodosa** | * | * | * | * | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. grossheimi** | * | * | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. lateriflora** | * | * | * | * | * | * | * | | | | | | | | | | | | | | | | | | | | | |
| **S. canina** | * | * | * | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. ningpoensis** | * | * | * | * | * | * | * | * | | | | | | | | | | | | | | | | | | | | |
| **S. ilwensis** | * | * | * | * | * | * | * | * | | | | | | | | | | | | | | | | | | | | |
| **S. scorodonia** | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * |
| **S. alata** | * | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. vernalis** | * | * | * | * | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. variegata** | * | * | * | * | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. chrisanta** | * | * | * | * | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. koraiensis** | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | |
| **S. huerggeriana** | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | |
| **S. spicata** | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | |
| **S. koeltii** | * | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| **S. scopolii** | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | |
| **S. leucocladan** | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | * | |

(1) Aucuboside
(2) Scrophularioside
(3) 10-O-β-glucosylaucuboside
(4) Bartsioside
(5) Scrofheanoside I
(6) Harpagide
(7) 8-O-acetyl-harpagide
(8) (p-Methoxycinnamoyl)-harpagide
(9) Harpagoside
(10) 8-O-cis-cinnamoyl-harpagide
(11) 8-O-ferfuloyl-harpagide
(12) 8-O-(2-hydroxy-cinnamoyl)-harpagide
(13) 6-O-α-L-galactopyranosyl-harpagide
(14) 6-O-α-L-rhamnopyranosyl-catalpol
(15) Koeltzioside
(16) Scropolioside A
(17) Scropolioside B
(18) Scropolioside D
(19) Scropioside B
(20) Scrofheanoside II
(21) 6-O-α-L-(2"-O-acetyl-3", 4"-O-di-p-methoxycinnamoyl-rhamno-pyranosyl)-catalpol
(22) Scrospioside B
(23) Saccatoside
(24) 6-O-α-L-(3"O-trans and cis-p-coumaroyl)-rhamnopyranosyl-catalpol
(25) Screovalentinioside
(26) Catalpol
(27) Picroside I
(28) 6-Methylcatalpol
(29) 6-O-(β-D-xylopiranosyl)-methyl catalpol
(30) Ajugol
(31) Laterioside
Iridoids from Scrophularia Genus


