Full Length Research Paper

Total tannin content in distinct *Quercus robur* L. galls

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The total tannin contents of three different *Quercus robur* L. galls collected from Estonia, three reference galls, and two plants were estimated. The dry common oak gall-apples tannin content was 47.2%; in brown shiny galls - 4.2%, and in grape galls - 3.4%; in Chinese galls - 89.1%, in Turkey galls - 81.4%, and in pistacia galls - 52.4%; in oak bark - 9.1%, and in common tormentil roots - 21.1%. The present study shows that the *Quercus robur* common oak gall-apples but not brown shiny galls or grape galls may be used as raw material for production of tannin containing traditional remedies.

Key words: Cynipid galls, *Quercus robur*, common oak, tannins.

INTRODUCTION

The history of traditional use of remedies derived from medicinal plants goes back to the ancient times. Due to distinct reasons - low cost for the source limited countries and search of alternatives for the industrialized world inhabitants - the utilization of medicines produced of medicinal plants is increasing (Li et al., 2009; Hall and Nazir, 2005; Seth and Sharma, 2004). As the chemical entities of the active pharmaceutical ingredients, among the herbal formulations the polyphenols and saponin derivatives prevail (Liu and Henkel, 2002). Tannins and related polyphenols have been implicated to various pharmacotherapeutic effects (Ferreira et al., 2008). In particular, the tannin containing remedies are in use as anthelmintics (Ketzi et al., 2006), antioxidants (Koleckar et al., 2008), antimicrobials and antivirals (Buzzini et al., 2008), and for the cancer treatment (Chung et al., 1998).

The tannins are widely distributed in almost all plant foods (Serrano et al., 2009; Manach et al., 2004). However, the primary source of tannins as active pharmaceutical ingredients are the medicinal plants while the polyphenolic compounds themselves are not always well characterized and there exist differences in polyphenol composition among various plant species (Okuda, 2005). In traditional medicine, the galls of various plants have been considered as tannin rich plant parts suitable for preparation of distinct polyphenol containing formulations (Allison and Schultz, 2005; Nyman and Julkunen-Titto, 2000; Lamien et al., 2005; Kaur et al., 2008; Vonshak et al., 2003). Galls are mainly used as a source of tannic acid, which is known as an astringent and styptic. Also galls are used for tanning and dyeing, and in the manufacture of inks (Evans, 2009).

The present study was intended to investigate the total tannin content in three different types of common oak galls collected from Estonia.

MATERIALS AND METHODS

Galls and plant samples

Three different types of common oak leaf (*Quercus robur* L., Fagaceae) galls were collected from Ahja Parish, Polvamaa County, Estonia. The oak leaves were infested by gall wasps (*Cynipidae*) as follows:

1) *Cynips* (Diplolepis) quercustoli L., referred as common oak gall-apples, Figure 1;
2) *Cynips* (Diplolepis) divisa Hart., referred as brown shiny galls;
3) *Neuroterus quercusbaccarum* L., referred as grape galls.

The common oak gall-apples, brown shiny galls, and grape galls were collected during the last decade of September that is the typical period for the beginning of oak defoliation in Estonia. For reference purpose, the samples of oak bark, common tormentil *Potentilla erecta* (L.) Raesusch rhizomatous roots, the Turkey gall (*Gallae Turcicae*), the Chinese galls (*Gallae Chinensis*), and pistacia galls (*Gallae pistaciae*) were also used. The oak bark and tormentil samples originated from the same area as the oak leaf.
galls while the Turkey, Chinese, and pistacia galls were from the pharmacognostic collection of the University of Tartu, Department of Pharmacy.

The galls and reference plant samples after cutting to pieces were dried during 10 days at room temperature in the absence of light in a well ventilated room. The air-dried samples were powdered in a mortar and powder sieved through 1 mm sieve. The relative moisture contents sample were: common oak gall-apples - 8.2%, brown shiny galls - 7.7%, grape galls - 4.6%, oak bark - 7.7%, common tormentil roots - 7.9%, Turkey galls - 8.1%, Chinese galls - 6.8%, and pistacia galls - 6.1%. All investigated galls or plant parts were in quadruplicate, the reported data values are adjusted for dry material and given as per cent average ± SD.

Voucher specimens of all galls and plant samples are deposited at the Department of Pharmacy, University of Tartu (Nooruse 1, Tartu 50411, Estonia).

**Estimation of tannin content**

Prior the quantitative estimation of the tannin content in the samples, the presence of tannins was identified using the classic FeCl₃ and Gelatin tests. The quantitative tannin content in samples was estimated by the method of Price and Butler (Price and Butler, 1977), with some modifications. In short, 0.1 g of a dry gall or plant sample was transferred to 100 ml flask; 50 ml water was added, and boiled for 30 min. After filtration with cotton filter the solution was further transferred to a 500 ml flask and water was added ad 500 ml mark. 0.5 ml aliquots were finally transferred to vials, 1 ml 1% K₂Fe(CN)₆ and 1 ml 1% FeCl₃ were added, and water was added ad 10 ml volume. After five min time period, the solutions were measured spectrophotometrically at 720 nm. The actual tannin concentrations were calculated on the basis of the optical absorbance values obtained for the standard solutions in range 5 - 25 µg /10 ml.

**Statistics**

The data for tannin contents of galls were analyzed using the one way analysis of variance (ANOVA), when necessary, the Dunnett’s post hoc test was applied (the data for the Chinese galls served as the control group). The reference values of tannin contents for oak bark and tormentil roots were excluded from ANOVA procedure.

**RESULTS AND DISCUSSION**

In the Northern Europe, the main source of daily tannin intake is the distinct berries and berry products (Heinonen, 2007). On the other hand, for medicinal purpose, in the Northern Europe the oak bark has been used as a source for the preparation of tannin containing traditional remedies. It is well documented that the oak bark is rich of tannins (Scalbert et al., 1988; Dury et al., 1998). This was also verified by the present study which demonstrates that the oak bark tannin content was nine per cent. We also confirmed that the common tormentil dry rhizomatous roots contain about 20 per cent tannins.

Though in the global scale, the Chinese galls are very popular raw materials for traditional medicine formulations (Liu and Henkel, 2002; Ikai and Hijii, 2007), the oak galls may be an alternative for the regions where traditional Chinese galls are not freely available (Kaur et al., 2008; Ikai and Hijii, 2007). The main result of our present study is the finding that the common oak gall-apples collected from Estonia contain about 47% tannins (Table 1) and thus, they can be considered as tannin rich plant parts. Though this value is statistically significantly lower as compared with the Chinese or Turkey galls, the absolute values of tannin content in common oak gall-apples are still sufficient to be used as raw material for the preparation of tannin containing remedies. Furthermore, we herewith also demonstrate that the brown shiny galls and grape galls of oak leaves do not possess perspectives for the wide use in traditional medicine what concerns the tannin containing formulations since their tannin content is even lower than in oak galls.
Table 1. Tannin contents in galls and reference plant samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Tannin content (% for dry material)</th>
</tr>
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<tbody>
<tr>
<td>Chinese galls</td>
<td>89.1±4.8</td>
</tr>
<tr>
<td>Common oak gall-apples</td>
<td>47.2±2.9***</td>
</tr>
<tr>
<td>Brown shiny galls</td>
<td>4.2±0.3***</td>
</tr>
<tr>
<td>Grape galls</td>
<td>3.4±0.4***</td>
</tr>
<tr>
<td>Turkey galls</td>
<td>81.4±4.8**</td>
</tr>
<tr>
<td>Pistacia galls</td>
<td>52.4±3.9***</td>
</tr>
<tr>
<td>Oak bark</td>
<td>9.1±0.8</td>
</tr>
<tr>
<td>Common tormentil roots</td>
<td>21.1±0.3</td>
</tr>
</tbody>
</table>

**p < 0.01; ***p < 0.001, significantly different as compared with Chinese galls, Dunnett’s test after a significant one way ANOVA effect was revealed [F(5,18) = 9.42, p < 0.001].

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