International Scholars Journals

International Journal of Enology and Viticulture Vol. 6 (6), pp. 001-004, June, 2019. Available online at www.internationalscholarsjournals.org © International Scholars Journals

Full Length Research Paper

Total tannin content in distinct *QUERCUS ROBUR* L. galls

Urve Paaver, Vallo Matto* and Ain Raal

Department of Pharmacy, University of Tartu, Nooruse 1, 50441 Tartu, Estonia.

Accepted 12 March, 2019

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 antihelmintics (Ketzis 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-Tiitto, 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.

*Corresponding author. E-mail: vallo.matto@ut.ee. Tel: +372 737 5197. Fax +372 737 5289.

MATERIALS AND METHODS

Galls and plant samples

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

1) *Cynips (Diplolepis) quercusfolii* 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.) Raeusch 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

Figure 1. A photographic image of common oak gall-apples collected from Ahja Parish, Pôlvamaa County, Estonia.

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 $FeCl_3$ 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% $K_3Fe(CN)_6$ and 1 ml 1% $FeCl_3$ 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

Table 1. Tannin contents in galls and reference plant samples.

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

**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 [$F(5,18) = 9.42$, $p < 0.001$].

bark. In addition, these galls are of smaller size and thereby laborious to collect. Nevertheless, the tannin contents of ca four per cent and ca three per cent for the common oak (*Quercus robur*) leaf galls inflicted by *Cynips (Diplolepis) divisa* and *Neuroterus quercusbaccarum*, respectively, are in good agreement with the data reported earlier studies (Hartley, 1998). The irrelevant differences of tannin contents among research reports can be explained by the facts that in oak galls the content of polyphenolic compounds depends also on the gall structure investigated (Allison and Schultz, 2005) as well as on the oak species (Taper and Case, 1987).

The microbiological effect of gall derived tannins is well described in the study of Vonshak et al., 2003. The authors found that an aqueous extract of galls of *Terminalia chebula* elicits inhibitory effect on three dermatophytes (*Trichophyton spp*.) and three yeasts (*Candida spp*.) while seeds extract of *T. chebula* inhibited only the growth of *T. glabrata* (Vonshak et al., 2003). This study clearly indicates that the plant gall derived remedies may have distinct pharmacotherapeutic effect as compared with analogues produced from other plant parts of the same plant species. Extrapolating the former idea into our study context, it is probable that the polyphenolic composition of the common oak galls and bark may be different, in particular with respect to the antimicrobial activity. Further studies discriminating the distinct forms of tannins using solvents based on different polarity will add supplementary information regarding this issue.

In sum, taking together the high tannin content of the common oak gall-apples and putatively distinct tannin composition, one can suggest that the common oak gall-apples of the *Quercus robur* leaves may be used as an alternative source for tannin containing traditional remedies.

ACKNOWLEDGEMENT

The authors thank Dr. Ass. Professor Mati Martin (Department of Zoology, University of Tartu) for identification of galls and pharmacist Ljubov Meshalkina for help at the laboratory.

REFERENCES

Allison SD, Schultz JC (2005). Biochemical responses of chestnut oak to a galling cynipid. J. Chem. Ecol. 31: 151-166.

Buzzini P, Arapitsas P, Goretti M, Branda E, Turchetti B, Pinelli P, Ieri F, Romani A (2008). Antimicrobial and antiviral activity of hydrolysable tannins. Mini. Rev. Med. Chem. 8: 1179-1187.

Chung KT, Wong TY, Wei CI, Huang YW, Lin Y (1998). Tannins and human health: A review. Crit. Rev. Food Sci. Nutr. 38: 421-464.

Dury SJ, Good JEG, Perrins CM, Buse A, Kaye T (1998). The effects of increasing CO_2 and temperature on oak leaf palatability and the implications for herbivorous insects. Global Change Biol. 4: 55-61.

Ferreira D, Gross GG, Hagerman AE, Kolodziej H, Yoshida T (2008). Tannins and related polyphenols: Perspectives on their chemistry, biology, ecological effects, and human health protection. Phytochemistry 69: 3006-3008.

Evans E (2009). Trease and Evans pharmacognosy. 16th edition. Saunders, Edinburg.

Hall M, Nazir N (2005). Quantification of the scientific research in the United States about popular herbal remedies. Altern. Ther. Health Med.11: 34-37.

Hartley SE (1998). The chemical composition of plant galls: Are levels nutrients and secondary compounds controlled by the gall-former? Oecologia 113: 492-501.

Heinonen M (2007). Antioxidant activity and antimicrobial effect of berry phenolics - a Finnish perspective. Mol. Nutr. Food Res. 51: 684-691.

Ikai N, Hijii N (2007). Manipulation of tannins in oaks by galling cynipids. J. For. Res. 12: 316-319.

Kaur G, Athar M, Alam MS (2008). Quercus infectoria galls possess antioxidant activity and abrogates oxidative stress-induced functional alterations in murine macrophages. Chem. Biol. Interact. 171: 272-282.

Ketzis JK, Vercruysse J, Stromberg BE, Larsen M, Athanasiadou S, Houdijk JG (2006). Evaluation of efficacy expectations for novel and non-chemical helminth control strategies in ruminants. Vet. Parasitol. 139: 321-335.

Koleckar V, Kubikova K, Rehakova Z, Kuca K, Jun D, Jahodar L, Opletal L (2008). Condensed and hydrolysable tannins as antioxidants influencing the health. Mini. Rev. Med. Chem. 8: 436-447.

Lamien CE, Meda A, Mans J, Romito M, Nacoulma OG, Viljoen GJ (2005). Inhibition of fowlpox virus by an aqueous acetone extract from galls of Guiera Senegalensis J. F. Gmel (Combretaceae). J. Ethnopharmacol. 96: 249-253.

Li HY, Cui L, Cui M (2009). Hot topics in Chinese herbal drugs research documented in PubMed/MEDLINE by authors inside China and

outside of China in the past 10 years: Based on co-word cluster analysis. J. Altern. Complement Med. 15: 779-785.

Liu J, Henkel T (2002). Traditional Chinese medicine (TCM): Are polyphenols and saponins the key ingredients triggering biological activities? Curr. Med. Chem. 9: 1483-1485.

Manach C, Scalbert A, Morand C, Remesy C, Jimenez L (2004). Polyphenols: Food sources and bioavailability. Am. J. Clin. Nutr. 79: 727-747.

Nyman T, Julkunen-Tiitto R (2000). Manipulation of the phenolic chemistry of willows by gall-inducing sawflies. Proc. Natl. Acad. Sci. U S A. 97: 13184-13187.

Okuda T (2005). Systematics and health effects of chemically distinct tannins in medicinal plants. Phytochemistry 66: 2012-2031.

Price ML, Butler LG (1977). Rapid visual estimation and spectrophotometric determination of tannin content of sorghum grain. J. Agric. Food Chem. 25: 1268-1273.

Scalbert A, Monties B, Favre JM (1988). Polyphenols of Quercus-robur - adult tree and in vitro grown galli and shoots. Phytochemistry 27: 3483-3488.

Serrano J, Puupponen-Pimia R, Dauer A, Aura AM, Saura-Calixto F (2009). Tannins: Current knowledge of food sources, intake, bioavailability and biological effects. Mol. Nutr. Food. Res. 53 Suppl. 2: S310-329.

Seth SD, Sharma B (2004). Medicinal plants in india. Indian J. Med. Res. 120: 9-11.

Taper ML, Case TJ (1987). Interactions between oak tannins and parasite community structure - unexpected benefits of tannins to cynipid gall-wasps. Oecologia. 71: 254-261.

Vonshak A, Barazani O, Sathiyamoorthy P, Shalev R, Vardy D, Golan-Goldhirsh A (2003). Screening South Indian medicinal plants for antifungal activity against cutaneous pathogens. Phytother. Res. 17: 1123-1125.