Food Chemistry 147 (2014) 367–376

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Analytical Methods

Centauries as underestimated food additives: Antioxidant and antimicrobial potential

Branislav Šiler, Suzana Živković, Tijana Banjanac, Jelena Cvetković, Jasmina Nestorović Živković, Ana Ćirić, Marina Soković, Danijela Mišić *

Institute for Biological Research "Siniša Stanković", University of Belgrade, Bulevar despota Stefana 142, 11060 Belgrade, Serbia

ARTICLE INFO

Article history:
Received 3 September 2012
Received in revised form 16 September 2013
Accepted 1 October 2013
Available online 11 October 2013

Keywords:
Centauries
Secoiridoid glycosides
Phenolics
Antimicrobial activity
Antioxidant activity

ABSTRACT

Methanol extracts of aerial parts and roots of five centaury species (*Centaurium erythraea*, *C. tenuiflorum*, *C. littorale* ssp. *uliginosum*, *C. pulchellum*, and *Schenkia spicata*) were analysed for their main secondary metabolites: secoiridoid glycosides, a group of monoterpenoid compounds, and phenolics (xanthones and flavonoids), and further investigated for antioxidant capacity and antimicrobial activity. The results of ABTS, DPPH, and FRAP assays showed that above ground parts generally displayed up to 13 times higher antioxidant activity compared to roots, which should be related to higher phenolics content, especially flavonoids, in green plant organs. Secoiridoid glycosides showed no antioxidant activity. All the tested extracts demonstrated appreciative antibacterial (0.05–0.5 mg ml^{-1}) and strong antifungal activity (0.1–0.6 mg ml^{-1}). Our results imply that above ground parts of all centaury species studied, could be recommended for human usage as a rich source of natural antioxidants and also in food industry as strong antimicrobial agents for food preservation.

1. Introduction

The protection of human body against oxidative stress and/or pathogenic bacteria and fungi relies not only on endogenous defense mechanisms, but also on exogenous compounds taken in food and beverages. Many naturally occurring compounds found in edible and medicinal plants and spices have been shown to possess strong antioxidant (Gülçin, 2012) and antimicrobial activity (Tajkarimi, Ibrahim, & Cliver, 2010) and could serve as a natural source of bioactive molecules which significantly support the defense against various diseases and disorders.

European centauries (genera *Centaurium* and *Schenkia*, fam. *Gentianaceae*) are of great importance for food and beverage bittering, and have been widely used since ancient times. Centauries, especially *Centaurium erythraea*, are popular constituents of many modern gastric herbal preparations and dietary supplements (e.g. Botion, Ferreira, Côrtes, Lemos, & Braga, 2005). This species is listed in the Council of Europe (EMA, 2009) as a natural source of food flavoring, in category N2 (Newall, Anderson, & Phillipson, 1996), which could be added to food stuffs in small quantities (0.0002–0.0008%). Other species investigated in this study are often referred as a substitute for *C. erythraea* in "Centaurii herba" drug (Bisset, 1994). Numerous pharmacological effects have been attributed to centauries, including stomachic, digestive, antiinflammatory and antipyretic effects (Berkan, Ustünes, Lermioglu, & Özer, 1991; Newall et al., 1996), cholagogue, hepatoprotective, gastroprotective (Tuluce, Ozkol, Koyuncu, & Ine, 2011), diuretic (Haloui, Louedec, Michel, & Lyoussi, 2000), wound-healing, antimicrobial (Ross et al., 2011; Šiler et al., 2010), and antioxidant activities (Shahat et al., 2003; Valentão et al., 2001).

Phytochemical studies of centauries revealed the presence of secoiridoid glycosides (SGs) (C-10 monoterpenoids, which are built of isoprenoid units), and phenolics (xanthones, phenolic acids and their derivatives) as main constituents (Beerhues & Berger, 1995; Bibi, Ali, Sadozai, & Atta-ur-Rahman, 2006; Valentão et al., 2002; van der Sluis, 1985). The most abundant SGs (swertiamarin (SM), gentiopicrin (GP), and sweroside (SW)) (Fig. 1) show a vast spectrum of biological activities, such as fungitoxic, antibacterial (Šiler et al., 2010), gastroprotective (Niiho et al., 2006), hepatoprotective (Kondo, Takano, & Hojo, 1994), sedative (Bhattacharya, Reddy, Ghosal, Singh, & Sharma, 1976), and antitumor (Ishiguro et al., 1988). There are many forms of xanthones (basic skeleton C6–C1–C6) isolated from centauries, including eustomin, demethyleustomin, decussatin, and methylbellidifolin (Fig. 1) (Beerhues & Berger, 1995; Jensen & Schripsema, 2002; Krstić, Janković, Šavikin-Fodulović, Menković, & Grubišić, 2003), and they also show a wide range of biological activities (Ross et al., 2011). Some flavonoids (basic skeleton C6–C3–C6) isolated from centaury species are reported to possess antioxidant potential (Shahat et al., 2003).

This work was aimed at investigating the antioxidant potential and antimicrobial activity of five centaury species: *C. erythraea*

* Corresponding author. Tel./fax: +381 11 2078 404.
E-mail address: dmisic@ibiss.bg.ac.rs (D. Mišić).

0308-8146/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.foodchem.2013.10.007

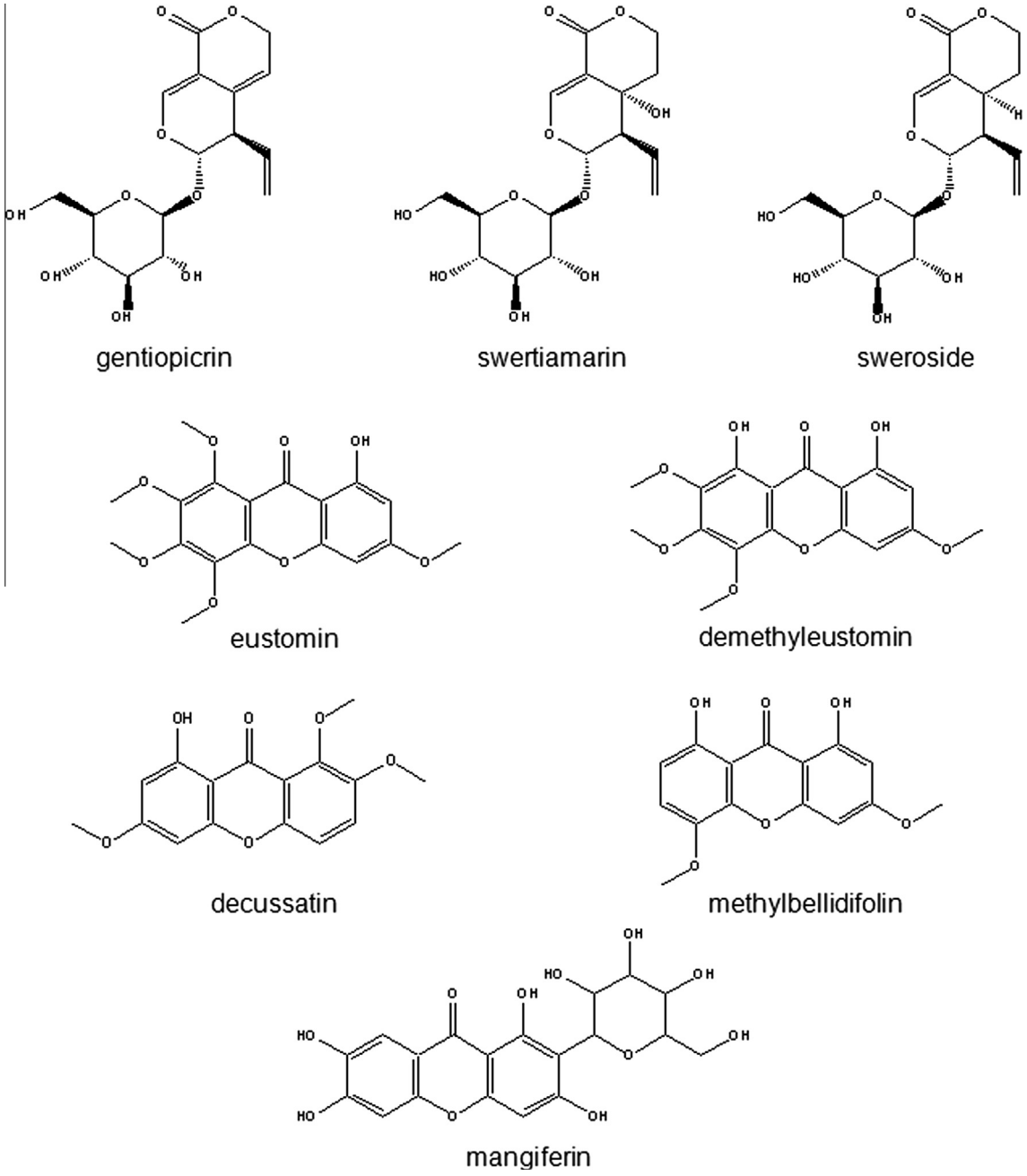

Fig. 1. Chemical structures of targeted secoiridoid glycosides (gentiopicrin, swertiamarin, and sweroside) and xanthones (eustomin, demethyleustomin, decussatin, methylbellidifolin, and mangiferin).

Rafn, *C. tenuiflorum* (Hoffmans. & Link) Fritsch, *C. littorale* (Turner) Gilmour ssp. *uliginosum* (Waldst. & Kit.) Melderis, *C. pulchellum* (Sw.) Druce, and *Schenkia spicata* (L.) Mansion, and to correlate the observed biological activities with the main constituents content detected in methanol extracts of their above ground parts and roots. One more objective was to re-evaluate the potential of centauries for implementation in food industry, since their role as food additives has been neglected in last decades.

2. Materials and methods

2.1. Chemicals and reagents

Formic acid and acetonitile (HPLC grade) were purchased from J. T. Baker (Deventer, The Netherlands), and methanol from AppliChem (Cheshire, USA). Milli-Q water was generated by deionization (Millipore, Billerica, USA). Gallic acid, rutin hydrate, p-coumaric acid, and ferulic acid (Sigma–Aldrich, Steinheim, Germany) were used as standards for spectrophotometric assays. Gentiopicrin (>90% purity, Roth, Karlsruhe, Germany), sweroside and swertiamarin (both 98% purity, Oskar Tropitzsch, Marktredwitz, Germany), and mangiferin (Sigma–Aldrich, Steinheim, Germany) were used as standards for HPLC–DAD and ESI-MS/MS analyses. 1,1-diphenyl-2-picrylhydrazil (DPPH), 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulphonate) (ABTS), ferric-2,4,6-tri(2-pyridil)-1,3,5-triazine (Fe^{3+}-TPTZ), and Folin–Ciocalteau reagent were purchased from Sigma–Aldrich (Steinheim, Germany). Streptomycin (Sigma–Aldrich, Steinheim, Germany) and commercial fungicide bifonazole (Srbolek, Belgrade, Serbia) were used as reference compounds in antimicrobial assays.