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Original research article

# Phenolic composition of fruits from different cultivars of red and black raspberries grown in Poland

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A B S T R A C T

Fruits of 11 varieties of *Rubus idaeus* and 1 variety of *Rubus occidentalis* cultivated in Poland, were analyzed in terms of phenolic composition. The qualitative and quantitative determination of phenolics was performed by means of HPLC-DAD coupled with ESI–MS detector. Quantitative determination of anthocyanins, revealed significant differences in the levels of major anthocyanins in most red raspberry varieties, of which cyanidin 3-*O*-sophoroside was the most prevalent one. Black raspberries displayed 4–11 times more anthocyanin content compared to red raspberries, with the dominating anthocyanins being cyanidin 3-*O*-rutinoside and cyanidin 3-*O*-xylosyl-rutinoside.

Sanguiin H-6 was the main ellagitannin identified in all varieties, with 'Beskid', 'Laszka', 'Polana' and 'Litacz' standing out as the richest sources of sanguiin H-6. Ellagic acid was at similar, low concentrations, with the highest levels observed in 'Willamette'. A number of other compounds belonging to flavonols, flavan-3-ols and phenolic acids, were found at much lower levels than anthocyanins and ellagitannins, with the presence of procyanidins $B_1$ and $B_2$ being reported for the first time in the fruits of black raspberries. Data obtained on the variation in content of major and minor phenolic compounds between raspberry varieties highlights the most valuable cultivars to be used as a dietary source of phenolic compounds.

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## 1. Introduction

The fruits of red raspberries (*Rubus idaeus*) and black raspberries (*Rubus occidentalis*) are popular food products and consumer demand for them is on the rise. They are recognized as a rich dietary source of phenolic compounds, which is linked to their health promoting and protective properties against a variety of human diseases (Gulcin et al., 2011; Nile and Park, 2014).

The two dominant groups of phenolics in raspberries are anthocyanins and ellagitannins (Mazur et al., 2014; Nile and Park, 2014). Several anthocyanins have been reported in red raspberries to date, predominantly cyanidin glucosides, sophorosides, rutinosides and glucosyl-rutinosides as well as low to trace levels of pelargonidin derivatives (Bradish et al., 2012; De Ancos et al., 1999; García-Viguera et al., 1998; Jakobek et al., 2009; Krauze-Baranowska et al., 2014; Lee et al., 2012; Määttä-Riihinen et al., 2004; Mazur et al., 2014; Mullen et al., 2002; Rao and Snyder, 2010; Sparzak et al., 2010). Less commonly reported anthocyanins comprise cyanidin 3-*O*-sambubioside and cyanidin 3-xylosyl-rutinoside (Krauze-Baranowska et al., 2014; Mullen et al., 2002). Reports about anthocyanin content in black raspberries are fewer in comparison (Dossett et al., 2010; Hecht et al., 2006; Krauze-Baranowska et al., 2014; Paudel et al., 2014; Tian et al., 2005; Tian et al., 2006; Wu and Prior, 2005; Wyzgoski et al., 2010), but the existing reports point out up to fivefold greater anthocyanin content (Kim et al., 2014; Krauze-Baranowska et al., 2014), and anthocyanin profile similar to that of red raspberries (Krauze-Baranowska et al., 2014; Lee et al., 2012) with the absence of cyanidin and peonidin glucosyl-rutinosides, pelargonidin 3-*O*-sophoroside and presence of peonidin 3-*O*-rutinoside instead (Lee et al., 2012). The two major elagitannins in *R. idaeus* comprise the dimeric HHDP (hexahydroxydiphenic) sanguiin H-6 and a tetrameric HHDP lambertianin C, as well as small amounts of ellagic acid, which is the product of their hydrolysis (Gulcin et al., 2011; Jakobek et al., 2009; Kähkönen et al., 2012; Krauze-Baranowska et al., 2014; Lee et al., 2012; Määttä-Riihinen et al., 2004; Mazur et al., 2014; Mullen et al., 2002; Rao and Snyder, 2010; Vuorela et al., 2005). Other elagitannins have also been reported, most notably sanguiin H-10, potentilin, casuarictin, galloyl- and galloyl-bis-HHDP-glucose (Kähkönen et al., 2012; Lee et al., 2012), as well

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as a number of ellagic acid conjugates in the form of pentosides, methyl pentosides and acetyl pentosides (Lee et al., 2012; Määttä-Riihinen et al., 2004; Mullen et al., 2002). To the best of our knowledge there is only one report outlining the content of sanguiin H-6 and ellagic acid in *R. occidentalis* fruits (Krauze-Baranowska et al., 2014).

Other phenolics like flavonols (quercetin, kaempferol and their glycosides), flavan-3-ols (catechin, epicatechin), proanthocyanidins (procyanidin $B_2$) and a number of hydroxycinnamic acids (*p*-coumaric, ferulic, caffeic, gallic, sinapic, vanillic, cinnamic acids) and their conjugates, have also been reported in the fruits of the red raspberry (Gulcin et al., 2011; Jakobek et al., 2009; Lee et al., 2012; Määttä-Riihinen et al., 2004; Rao and Snyder, 2010), however they occur in significantly lower concentrations (Vuorela et al., 2005). Data on flavonols and phenolic acids present in black raspberries is lacking as well, with few reports on the presence of quercetin, *p*-coumaric, ferulic and caffeic acids (Lee et al., 2012).

Both ellagitannins and anthocyanins have been reported to possess numerous biological activities, such as antioxidant (Jeong et al., 2010; Kähkönen and Heinonen, 2003; Liu et al., 2002), anti-inflammatory (Jean-Gilles et al., 2012; Raudone et al., 2014; Sangiovanni et al., 2013) and antimicrobial (Heinonen, 2007; Krauze-Baranowska et al., 2014), while anthocyanin-rich black raspberries have been shown to exhibit chemopreventive and anticancer properties (Hsieh et al., 2013; Johnson et al., 2011; Kula and Krauze-Baranowska, 2016). Flavonols, flavan-3-ols, proanthocyanidins and phenolic acids, although present in much lower amounts, can be an active contributor to the biological effects of raspberry fruits (Kumar and Pandey, 2013) and should also be taken into consideration in phytochemical investigations.

As the public becomes more aware of the health implications of diets, consumer interest in food products rich in polyphenols is on the rise. Therefore investigating composition variation between raspberry cultivars could provide valuable information to identify varieties with the highest levels of beneficial phenolic compounds.

There are many factors that can influence the content of phenolic compounds in raspberry fruits, including genetic, environmental factors and seasonal variation (Anttonen and Karjalainen, 2005; Kassim et al., 2009; Mazur et al., 2014). The current evidence shows that phenolic content can vary significantly between cultivars (Anttonen and Karjalainen, 2005; Gulcin et al., 2011; Krauze-Baranowska et al., 2014; Mazur et al., 2014; Stewart et al., 2007), and although some interest has been given to the chemical composition of red and black raspberry fruits, detailed information about their variation in cultivars is still scarce.

The aims of the present study was quantitative and qualitative comparison of anthocyanin and ellagitannin profiles in the fruits of several cultivars of red raspberry (*R. idaeus*) and black raspberry (*R. occidentalis*) grown in Poland, paired with an investigation of flavonols, flavan-3-ols, proanthocyanidins and phenolic acids in the analyzed plant material.

## 2. Materials and methods

### 2.1. Chemicals

Standard compounds of gallic acid, caffeic acid, ellagic acid, chlorogenic acid, salicylic acid, kaempferol, quercetin, myricetin, hyperoside, isoquercetin, kaempferol 3-*O*-glucuronide and catechin were obtained from Fluka (Switzerland). Protocatechuic acid,

**Table 1**
Description of *R. idaeus* and *R. occidentalis* varieties.

| Variety | Pedigree | Developed in | Harvest dates |
|---|---|---|---|
| *R. idaeus* 'Benefis' | (Beskid × Tulameen) | Poland, 2006 | 20 July 2009<br>9 July 2010<br>16 July 2010 |
| *R. idaeus* 'Beskid' | (Malling Landmarka × Preussen × New- Burgh × Chief × Cumberland × *Rubus cockburnianus*) | Poland, 1991 | 17 July 2009<br>22 July 2009<br>16 July 2010 |
| *R. idaeus* 'Glen Ample' | Complex parentage, includes Glen Prosen and Meeker | Scotland | 29 July 2009<br>9 August 2009<br>6 August 2009 |
| *R. idaeus* 'Heritage' | (Milton × Cuthbert × Durham) | USA, 1969 | 19 August 2009<br>23 August 2009<br>15 September 2010 |
| *R. idaeus* 'Laszka' | | Poland, 2006 | 20 July<br>2 June 2010<br>2 July 2010 |
| *R. idaeus* 'Pokusa' | (Autumn Bliss × Heritage × *Rubus odoratus* × *Rubus occidentalis*) | Poland, 2003 | 23 August 2009<br>27 August 2010<br>3 September 2010 |
| *R. idaeus* 'Polana' | (Heritage × Zeva Herbsternte) | Poland, 1991 | 19 August 2009<br>26 August 2009<br>27 August 2010 |
| *R. idaeus* 'Polesie' | (Autumn Bliss × Heritage) | Poland, 2006 | 12 August 2009<br>15 September 2009<br>15 September 2010 |
| *R. idaeus* 'Polka' | | Poland, 2003 | 12 August 2009<br>15 September 2009<br>15 September 2010 |
| *R. idaeus* 'Poranna Rosa' | (Polana × *Rubus occidentalis* × *Rubus coreanus*) | Poland, 2002 | 23 August 2009<br>3 September 2009<br>15 September 2010 |
| *R. idaeus* 'Willamette' | | USA, 1943 | 2 July 2009<br>12 August 2009<br>28 September 2010 |
| *R. occidentalis* 'Litacz' | Bristol | Poland, 2008 | 20 July 2009<br>09 July 2010<br>16 July 2010 |

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