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Quality and authenticity of the forest fruits through antioxidant compounds – a review on chemometric tools

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Abstract

The review focuses on the chemometric tools used for evaluation of the quality and authenticity of forest fruits (berries) through the antioxidant compounds analysis and fingerprinting. The classification of forest fruits, especially growing in Romania, the composition and analysis techniques for the antioxidant compounds, as well as the "key" compounds have been presented. The coupling of the analysis techniques for the forest fruit antioxidants with chemometric tools is emphasized. The unsupervised and supervised pattern recognition techniques such as cluster analysis, principal component analysis, discriminant analysis and artificial neural networks have been considered for the quality and authenticity evaluation. Finally, the appropriate antioxidant compound analysis – chemometry coupling techniques were emphasized for the evaluation of the quality and authenticity of autochthonous forest fruits.

Keywords: forest fruits, berries, quality and authenticity, fingerprint, chemometry, multivariate statistical analysis, cluster analysis, principal component analysis, discriminant analysis, coupling techniques

1. Forest fruits - berries

Forest fruits are berries that belongs to forest trees. They are edible fruits, but some of them can be poisonous. Berries such as elderberry are poisonous only when unripe. Berries grow even in wild regions or in crops. The most known are [1-3]: strawberry (e.g. *Fragaria* × *ananassa* Duchesne), raspberry (e.g. *Rubus idaeus* L.), blackberry (e.g. *Rubus fruticosus* L.), blueberry (e.g. *Vaccinium myrtillus* L.), cranberry (e.g. *Vaccinium oxycoccos* L.), gooseberry or currant (e.g. *Ribes rubrum* L.), mulberry (e.g. *Morus alba* L.), elderberry (e.g., *Sambucus ebulus* L.), dog rose (*Rosa canina* L.), sea buckthorn (*Hippophae rhamnoides* L.) etc.

More than six million tones are cultivated worldwide, the most important being strawberries (more than a half), red and black currants, gooseberries, blueberries, raspberries, cranberries, and blackberries [4]. Europe and North America have the highest productivity on strawberries, while for blueberries North America produces more than three quarters of the worldwide production. Almost a half of the raspberry's productivity is in Europe, while more than a third of the blackberry's productivity is in North America. Red and black currants are mainly produced in Asia and Europe.

2. Chemical composition of berries

The basic composition of berries comprise of carbohydrates, organic acids, enzymes, cell wall components, vitamins, pigments and minerals [5]. The presence of carbohydrates provide their sweetness, organic acids provide sour taste, while pigments reveal the characteristic color of the fruits. The specific fruity flavors are provided by the

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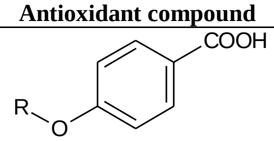
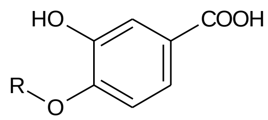
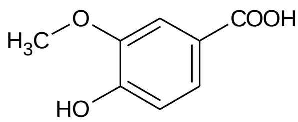
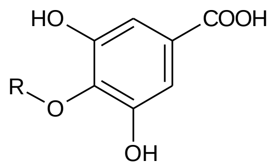
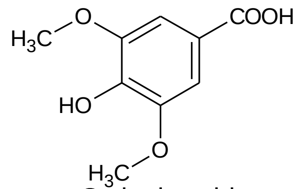
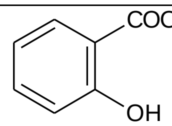
essential oil components and the overall aroma is generated by a combination of all senses. On the other hand, the presence of vitamins and polyphenolic pigments make these products very important for the human health. For example, approximate contents of various components in fresh blueberries are: carbohydrates 14.5%, total sugars 9.96% (mainly consisting of glucose 4.88% and fructose 4.97%), total fibers 2.4%, protein 0.74%, total lipids 0.33%, ash 0.24% and water 84.2%. Another example is for strawberry, with the following values: carbohydrates 7.68%, total sugars 4.66% (mainly consisting of glucose 2.04% and fructose 2.50%), total fibers 2.0%, protein 0.67%, total lipids 0.30%, ash 0.40% and water 90.95% [6, 7]. Among carbohydrates, berries contain water soluble sugars, especially glucose, fructose, and even sucrose, but also starch (amylose and amylopectin). The most important organic acids in berries are citric and malic acids, but phenolic acids can occur. The latest provide bitter and astringent characteristics. Enzymes are sometimes responsible for changing in organoleptic properties of berries when they are damaged, for example for obtaining puree. They belong to hydrolase and oxidase classes, such as polyphenol oxidase (responsible for browning), peroxidase, or polygalactouronase (cleavage of the pectins and influences the berry texture). Pectins, cellulose, hemicellulose, glycoproteins and polyphenol esters are the main components of the cell walls [7].

The most important berry constituents are anthocyanins, which provide various colors to these fruits, such as deep red, purple or blue [8]. Moreover, carotenoids are also an important, with β-carotene and lutein the main components from this class. Other berries' components having antioxidant properties are hydroxycinnamic acids and flavonoids [5]. Ascorbic acid (vitamin C) is also an antioxidant component, belonging to vitamin class. These antioxidant compounds are presented in the next section. Berries contain other vitamins such as B vitamins (niacin), folate, tocopherols and vitamin K. Regarding the minerals, calcium, magnesium, potassium and phosphorous are the most important [5].

3. Antioxidant compounds in berries

There are many classes of compounds having antioxidant activities. Most of them have one or more OH phenolic groups. These hydroxyl groups can occur in a free form in the antioxidant structure. On the other hand, they can be substituted by mono- and disaccharide moieties or by methyl group. In the first case, the ethereal bond can be easily spitted to the free hydroxyl group. Antioxidant compounds in berry fruits belong to phenolic acid, flavonol, flavonone, flavanol, and anthocyanidin classes, some of them being glycoside-based derivatives (e.g., flavonoid glycosides and anthocyanins) [5, 9-12]. A survey on the antioxidant compounds found in berry fruits is presented in Tables 1-6.

Table 1. The main antioxidant compounds identified in berry fruits – phenolic acids / benzoic acid derivatives

Antioxidant compound	Berry fruit
 p-Hydroxybenzoic acid derivatives	strawberry (R: Glu) blueberry (R: H or R: Glu) cranberry (R: H) raspberry (R: H)
 Protocatechuic acid derivatives	strawberry (R: H) blueberry (R: Glu)
 Vanillic acid derivatives	strawberry blueberry cranberry raspberry
 Gallic acid derivatives	blueberry (R: H or R: Glu) sea buckthorn (R: H) raspberry (R: H)
 Syringic acid derivatives	blueberry
 Salicylic acid	raspberry

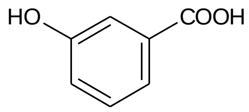
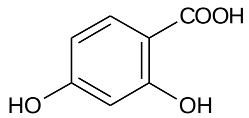
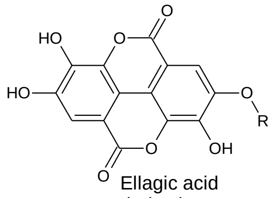
 3-Hydroxy-benzoic acid	cranberry
 2,4-Dihydroxy-benzoic acid	cranberry
 Ellagic acid derivatives	strawberry (R: Glu) blueberry (R: H) sea buckthorn (R: H)

Table 2. The main antioxidant compounds identified in berry fruits – phenolic acids / quinic acid derivatives

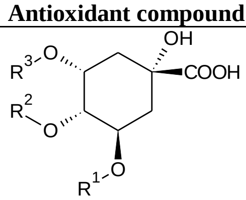
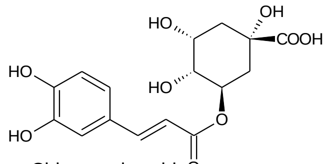
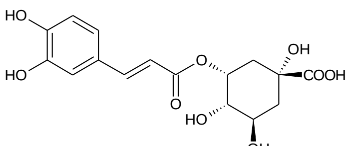
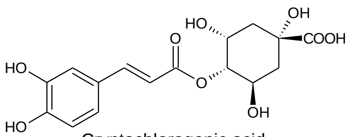
Antioxidant compound	Berry fruit
 Quinic acid derivatives	strawberry - derivatives - $R^{1,2}$: H, R^3: galloyl - $R^{1,2}$: H, R^3: caffeoyl - $R^{1,2}$: H, R^3: Glu blueberry (R^1: Glu, $R^{2,3}$: H or R^2: Glu, $R^{1,3}$: H or R^3: Glu, $R^{1,2}$: H or R^3: feruloyl, $R^{1,2}$: H or R^3: p-cumaroyl, $R^{1,2}$: H)
 Chlorogenic acid	blueberry (bilberry) elderberry
 Neochlorogenic acid	blueberry cranberry elderberry
 Cryptochlorogenic acid	elderberry

Table 3. The main antioxidant compounds identified in berry fruits – phenolic acids / cinnamic acid derivatives

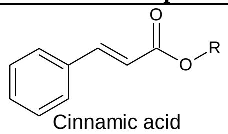
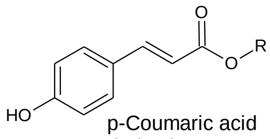
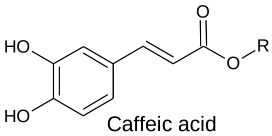
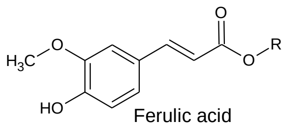
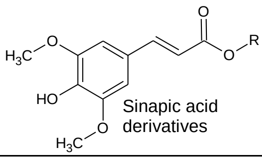
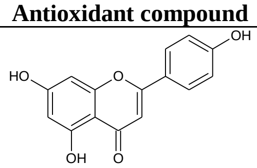
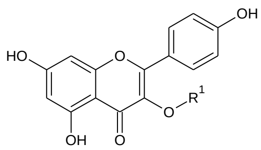
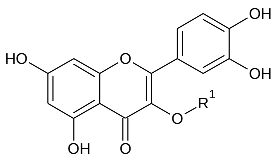
Antioxidant compound	Berry fruit
 Cinnamic acid derivatives	strawberry (R: H) bilberry (R: H) cranberry (R: H) raspberry (R: H)
 p-Coumaric acid derivatives	strawberry (R: H or 4-*O*-Glu) blueberry (R: H or 4-*O*-Glu) cranberry (R: H) sea buckthorn (R: H) raspberry (R: H) elderberry (R: H)
 Caffeic acid derivatives	blueberry (R: H or 4-*O*-Glu) bilberry (R: H) cranberry (R: H) raspberry (R: H) elderberry (R: H)
 Ferulic acid derivatives	strawberry (4-*O*-Glu) blueberry (R: H or 4-*O*-Glu) cranberry (R: H) sea buckthorn (R: H) raspberry (R: H)
 Sinapic acid derivatives	cranberry (R: H)

Table 4. The main antioxidant compounds identified in berry fruits – flavonoids / flavonols and flavonones

Antioxidant compound	Berry fruit
 Apigenin	raspberry
 Kaempferol & derivatives	strawberry (R^1: H, Glu or glucuronide) blueberry (R: H or Glu) cranberry (R: Glu) sea buckthorn (R: H) elderberry (R: H or Rut)
 Quercetin & derivatives	strawberry (R^1: H or glucuronide) blueberry (R: H or Glu or Ara or Gal or Xyl) cranberry (R: H or Ara or Gal or Rha) sea buckthorn (R: H) raspberry (R: glucuronide or Rut or Rha) elderberry (R: H or Glu or Rut)

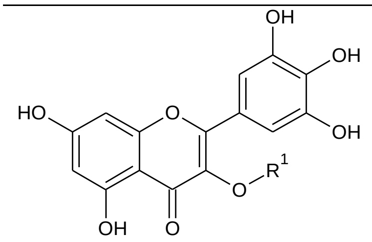
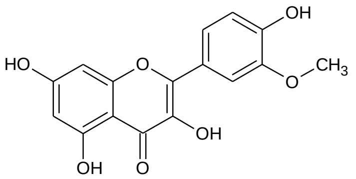
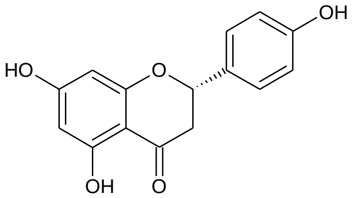
 Myricetin & derivatives	strawberry (R: H) blueberry (R: H or Ara or Rha) cranberry (R: H or Ara) elderberry (R: Rut)
 Isorhamnetin	sea buckthorn elderberry (R: H or Glu)
 Naringenin	raspberry

Table 5. The main antioxidant compounds identified in berry fruits – flavonoids / flavanols

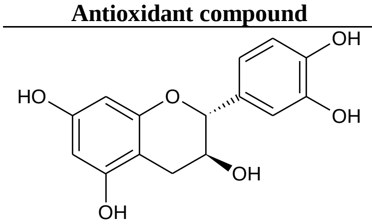
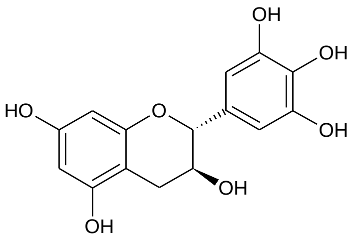
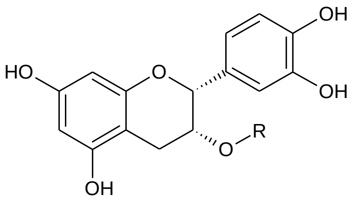
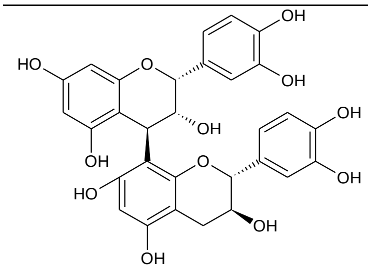
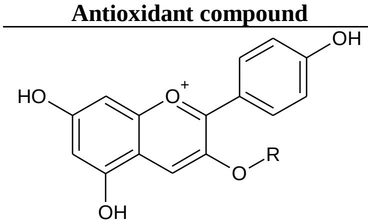
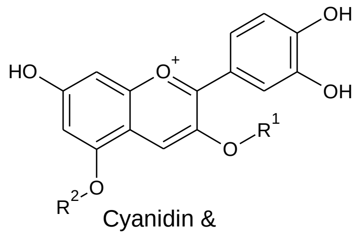
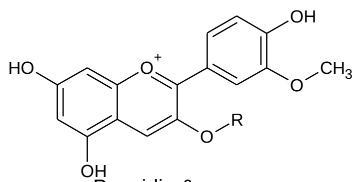
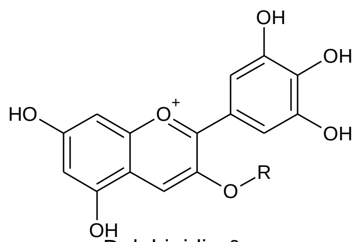
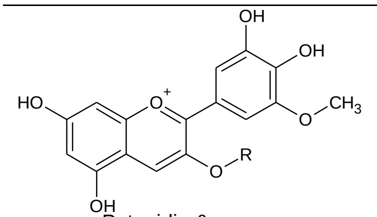
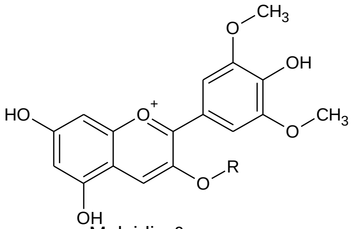
Antioxidant compound	Berry fruit
 (+)-Catechin	strawberry sea buckthorn raspberry elderberry
 (+)-Gallocatechin	strawberry
 (-)-Epicatechin & derivatives	strawberry (R: galloyl) blueberry (R: H) sea buckthorn (R: galloyl) elderberry (R: H)
 Procyanidin B1	strawberry sea buckthorn elderberry

Table 6. The main antioxidant compounds identified in berry fruits – anthocyanidins and anthocyanins

Antioxidant compound	Berry fruit
 Pelargonidin & derivatives	strawberry ($R^{1,2}$: H or R^1: malonyl-Glu, Ara, Glu, Rut, malyl-Glu and R^2:H) cranberry (R: Glu or Rut) elderberry (R^1: Glu or sambubioside, R^2: H)
 Cyanidin & derivatives	strawberry (R^1: succinyl-Glu, R^2: H or $R^{1,2}$: H) blueberry (R^1: Glu or Ara or Gal or 6-Ac-Glu/Gal, R^2: H) cranberry (R^1: Glu or Ara or Gal or sophoroside, R^2: H) raspberry (R^1: Glu or Rut or sophoroside or sambubioside or Glu-Rut or Xyl-Rut, R^2: H) elderberry (R^1: Glu or Rut or sambubioside, R^2: H; $R^{1,2}$: Glu)
 Peonidin & derivatives	blueberry (R^1: Glu or Ara or Gal or 6-Ac-Glu/Gal, R^2: H) cranberry (R: Glu or Ara or Gal)
 Delphinidin & derivatives	blueberry (R^1: Glu or Ara or Gal or 6-Ac-Glu/Gal, R^2: H) elderberry (R^1: Rut, R^2: H)

 Petunidin & derivatives	blueberry (R^1: Glu or Ara or Gal or 6-Ac-Gal, R^2: H) elderberry (R^1: Rut, R^2: H)
 Malvidin & derivatives	blueberry (R^1: Glu or Ara or Gal or 6-Ac-Glu, R^2: H)

4. Chemometric approaches on the quality and authenticity of the forest fruits - berries through antioxidant compounds

Chemometrics allows to extract significant information from a large database. Many of these methods belong to multivariate statistical analysis. It is the case of principal component analysis (PCA), partial least squares (PLS), factor analysis (FA), correspondence analysis (CA), (linear) discriminant analysis (LDA), hierarchical cluster analysis (HCA), or artificial neural networks (ANN). A review on the use of chemometry to evaluate de quality and authenticity of forest fruits – berries is presented in Table 7.

Table 7. Chemometric tools applied for the evaluation of the quality and authenticity of forest fruits – berries

Berry fruit	Chemometric tools	References
Aronia melanocarpa	PLS	[13]
Cornus mas	PCA	[9, 14, 15]
Fragaria species	PCA	[10]
Fragaria × ananassa	PCA, HCA, CA, PLS	[10, 16-23]
Fragaria vesca	PCA, HCA	[24]
Hippophae rhamnoides	PCA	[25, 26]
Morus alba	HCA, CA, PCA	[27-31]
Ribes species	PCA, HCA	[11]
Ribes rubrum	PCA, HCA, LDA	[32, 33]
Rosa canina	PLS	[13]
Rubus fruticosus	PCA, LDA	[34]
Rubus idaeus	PCA, HCA	[18, 35, 36]
Vaccinium species	HCA	[12]
Vaccinium angustifolium	PCA	[8, 37]
Vaccinium corymbosum	PCA	[8, 38, 39]
Vaccinium myrtillus	PCA, CA, LDA, HCA	[18, 32-34, 40]
Vaccinium oxycoccos	PCA, HCA	[18, 33, 41]
Vaccinium uliginosum	PCA	[38]

5. Conclusion

In this review the recent applications of chemometric tools on the discrimination, evaluation of the quality and adulteration of various forest fruits – berries have been emphasized. Among these, principal component analysis and hierarchical cluster analysis (PCA and HCA) have extensively used. The review was focused on berry fruits growing in Europe, as well as in Romania. The chemical composition, focused on antioxidant compounds, were also systematically presented, based on the main antioxidant compound classes. They were phenolic acids (as benzoic, quinic, cinnamic acid derivatives), flavonoids and anthocyanins. This recent survey can trigger the useful application of such chemometric tools for quality evaluation of forest fruit-based food products, including functional and innovative foods.

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