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HEALTH PROMOTING QUALITY OF THE ROMANIAN HONEY

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Abstract

Honey is a nutraceutical and its price is related to consumer perception on the health promoting quality. Last years, in the European Union, Romania was constantly present in the top three honey producers. The honey bee foraging areas from Romania support production of high-quality honey, because of their large biodiversity. The overlapping biogeography of Romania, which connects Central Europe, South-East-Europe and Eastern Europe is associated with a diversity of the agro-pedo-climatic condition, leading to large diversity of plant species producing nectar and pollen. A large area of meadows, still traditionally managed, offer further support for the biodiversity and honey bee foraging. Romania is on the top of the EU producers for entomophile oleaginous crops, rapeseed and sunflower. Forests cover more than one third of Romania, and they include Acacia and Tilia species, and produce large amount of polyphenol rich honeydew. However, the perception of the honey quality by foreign customers is still not in accordance with the Romanian honey quality. The aim of this paper is to review the published data regarding health promoting quality indicators of the Romanian honey, total polyphenols and flavonoids content, antioxidant and anti-bacterial activities, and to discuss the actions which are necessary in order to transform the biological activity into a higher profitability of the Romanian honey value chain.

Key words*: antioxidant activity, antibacterial activity, health promoting quality, honey, polyphenols.*

INTRODUCTION

Honey is a nutraceutical and its consumption and price are driven by the perception of its health and wellness promoting characteristics (Ajibola et al., 2012). The combination of the bioactive phytochemicals, harvested and concentrated by honeybee workers, and honeybee active proteins, released during honey formation, such as glucose oxidase (GOX) or defensin-1 (Def-1), were considered as being involved in the biological activity of honey (Kwakman et al., 2011). A special case of phytochemical is the methylglyoxal from New Zealand manuka honey. This type of honey is characterized for its antimicrobial effects and has a high price, proportionally to its biological effect, expressed as UMF (Unique Manuka Factor), which is associated with methylglyoxal (Adams et al., 2008; Mavric et al., 2008). The methylglyoxal was demonstrated to originate from the dihydroxyacetone, which is present in the nectar of manuka flowers Manuka honey, *Leptospermum scoparium* (Adams et al., 2009).

The honey bee foraging areas from Romania support production of high-quality honey (Juan-Borrás et al., 2014). Romania has a large biodiversity. The overlapping biogeography of Romania, which connects Central Europe, South-East-Europe and Eastern Europe is combined with a landscape which supports a mosaics of plant species, due to the heterogeneity of temperature and water supply (Sutcliffe et al., 2016). A large area of meadows, still traditionally managed (Dahlstrom et al., 2013), and low-intensity farming, further support biodiversity (Loos et al., 2014) and honey bee foraging. Such biodiversity, combined with heterogeneity of microclimatic conditions promotes bioactives accumulation (Oancea et al., 2013). The perennial weedy species, from meadows and step-asides areas, especially those of *Lamiaceae* and *Boraginaceae*, provide significant amount of nectars and phytochemicals, and offer opportunities for sustainable development of beekeeping and honey production (Ion et al., 2018).

Romania is on the top of the EU producers for entomophile oleaginous crops, rapeseed (Vasile et al., 2016) and sunflower (Turek Rahoveanu et al., 2018). Despite its low fructose/glucose ratio, which affects honey manipulation and storage, rapeseed honey contains polyphenols with a high biological value (Samborska et al., 2019). Sunflower honey, appreciated for its delicate flavor, has also a high content of antioxidants (Ozcan et al., 2017).

Forests cover more than one third of Romania and have a biomass production potential above European average (Neumann et al., 2016). The melliferous potential of the silver linden tree (*Tilia tomentosa*) growing in the South of Romania average 318 $kg.ha^{-1}$ (Ion et al., 2018). Acacia tree (*Robinia pseudoaccacia*) is considered the most valuable resource of the forests from South-East Romania (Vlad et al., 2012). Honeydew honey samples from North-East (Suceava) and North-West (Bihor) parts of Romania were proven to have a high content on valuable polyphenols, such as pinocembrin, chrysin, p-coumaric acid or myrecitin (Chis et al., 2016; Oroian et al., 2016).

Romania is one of the largest producers of honey from the European Union (Pippinato et al., 2019). In the last years, exports significantly exceeded imports (Isopescu et al., 2018). However, this positive trade balance is rather associated with a price lower than the average EU price for honey (Popescu et al., 2018; Terin et al., 2018). Therefore, the profitability of the whole Romanian honey value chain is rather low, despite the fact that Romanian apiculture needs better profitability, to support sustainability and resilience.

Beekeeping in Romania faces even more challenges than in other countries. Rainfed crops are predominant in Romanian agriculture. Such crop structure, combined with a increase risks of severe droughts, amplified by climate changes, could significantly affect agricultural yield (Trnka et al., 2010), including nectar and phytochemicals production. Aridity index increased in the last decades for fertile regions of Romania - Dobrogea and Bărăgan (Paltineanu et al., 2007), Oltenia Plain (Prăvălie et al., 2013). Only on Oltenia Plain (South-West of Romania) the area with aridization risk exceeds 700,000 hectares (Prăvălie et al., 2013).

Romanian forest capacity to generate renewable resources is influenced by relatively high share of illegal logging - which resulted also after changing in forest ownership and exploitation management (Bouriaud et al., 2016), legal management rules, which forbid tending and thinning (Bouriaud et al., 2016) and legacies of historical management (Munteanu et al., 2016). The melliferous potential of the main nectar producing trees, the silver linden tree and acacia, is affected not only by such factors, but also by climatic changes, especially drought (Vlad et al., 2012; Ion et al., 2018).

The aim of this paper is to review the published data regarding health promoting quality indicators of Romanian honey, total polyphenols, and flavonoids content, individual phenolics, antioxidants, anti-bacterial activities, and to discuss the actions which are necessary in order to transform the biological activity into a higher profitability of the Romanian honey value chain.

MATERIALS AND METHODS

Data regarding honey production in Romania in the last 6 years (total production, honey production, honey consumption per capita) and competitiveness of Romanian honey production (imported and exported quantities, trade balance index, price of honey on Romanian market and export price) were obtained from Eurostat, International Trade Center, FaoStat and National Institute for Statistics. The data obtained were compared with those from recently published papers (Isopescu et al., 2017; Popescu et al., 2018; Terin et al., 2018).

Data regarding Romanian honey composition and effects on human health were obtained from academic bibliographic database (ISI Web of Science Clarivate Analytics, Google Scholars, Scopus) using “polyphenols”, “honey”, “health effects” and “Romania” as the main keywords for queries.

The analysis was focused on polyphenols (phenolic acids, flavonoids, polycyclic polyphenols), because the main honey phytochemicals generally accepted as being involved in health effects on humans are polyphenols (Bogdanov et al., 2008; Cianciosi et al., 2018; Hossen et al., 2017; Hashim et al., 2021; Martinello et al., 2021).

RESULTS AND DISCUSSIONS

In the last 5 years, Romania was constantly among the top 3 producers of honey from the European Union (European Commission, 2020). The Romanian honey exports were almost constant, despite the fact that the level of honey productions has varied, mainly due to the variable climatic conditions, and the domestic consumption of honey significantly increased - Figure 1.

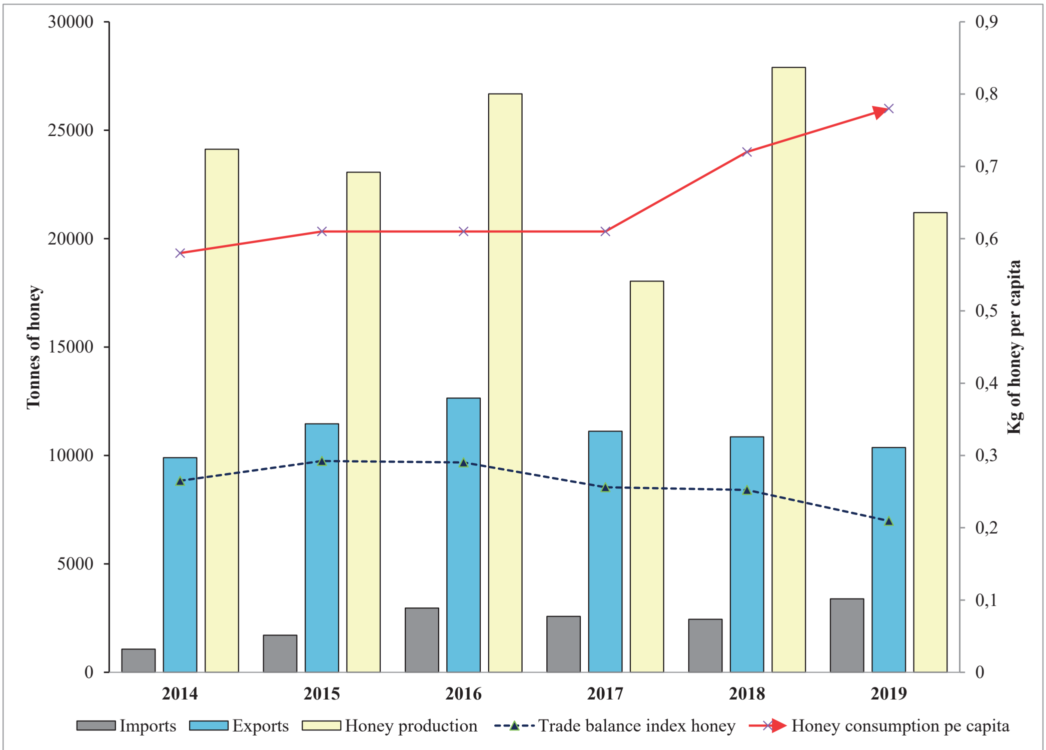

Figure 1. Evolution of the honey production, import, export, domestic consumption and trade index in the last years in Romania

These levels of production and the market dynamics are important also for crop pollination (Panța et al., 2018) and need more support from field crop farmers.

The main export destination for Romanian honey are EU countries. Together, Germany and Italy account for more than 50% of the Romania honey export/intra-community trade - Table 1. The highest price for Romanian honey is obtained in Japan. Honey export to Japan showed a decreased tendency in the last years, despite the fact that it is the most profitable destination. The tendency is toward markets with lower price - Figure 2.

These evolutions are determined by the main weakness of the Romania apiculture sector, large number of beekeepers, small apiary size, with a rather low honey yield (Grigoras et al., 2018). Despite the improvement of economic indicators related to specific number of beekeepers, beehives, and yield per bee hive, Romanian apiculture still faces this issue of fragmented production (Popescu et al., 2020). Due to this fragmentation, Romanian traders must spend money and energy to face challenges of honey authentication from a large numbers of suppliers. The analytical techniques required for honey authentication are sophisticated and expensive (Tsagkaris et al., 2021)

Table 1. The main destination of Romanian honey export

Export destination	% of Export 2019	Average price	Price evolution 2019 vs average price 2015-2019
World	***100%***	***€ 3,76***	***5%***
Germany	37%	€ 3,61	5%
Italy	17%	€ 3,70	4%
Japan	8%	€ 5,54	-17%
Denmark	4%	€ 3,86	16%
France	8%	€ 3,90	20%
Spain	6%	€ 3,97	10%
Poland	5%	€ 3,47	-7%
Austria	4%	€ 3,30	4%
United Kingdom	4%	€ 3,68	2%
Belgium	1%	€ 3,06	-11%
Others	5%	€ 4,08	-27%

Source: www.trademap.org, accessed at 21.04.2021

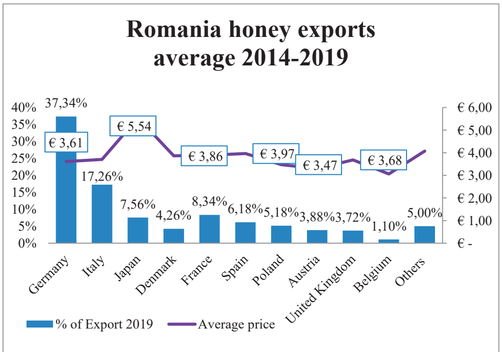

Figure 2. The evolution of the Romanian honey export price. Source: www.trademap.org, accessed at 21.04.2021

In order to better valorize Romanian honey, it is important to emphasize its quality. The willingness-to-pay a higher price was demonstrated to be related to the perception of the high-quality and not to other sustainability indicators such as organic certified production or biodegradable packaging (Bissinger et al., 2021).

Therefore, efforts are needed to demonstrate the quality of Romanian honey. Because honey is a nutraceutical, its quality must be related to the biological properties such as the antioxidant and antimicrobial activity, which are further correlated with honey bioactives, especially polyphenols.

There are several examples of honey which reached high price after scientific publications of papers which demonstrated their beneficial effects for human health. We already mentioned manuka honey and its UMF (Unique Manuka Factor), which is associated with methylglyoxal and the antibacterial properties of this honeydew honey (Adams et al., 2009).

Another example of expensive honey, due to its effects on human health, is the Mediterranean bitter honey, originating from strawberry-tree (*Arbutus unedo* L.). This type of honey has a high content of arbutin and polyphenols (Oses et al., 2020). Arbutin undergoes *in vivo* a hydrolysis to its aglycone, which is a hydroquinone, i.e., an oxidized polyphenol. Therefore, the antibacterial and anti-oxidant activity of honey could be related mainly to its polyphenols content.

The polyphenols content, in relation to the specific colloidal structure of honey, which generates a crowded space of the aqueous pockets. In this crowded water pocket polyphenols interact with honey specific proteins, glucose-oxidase (GOX) or defensin-1 (Def-1). This interaction is believed to be essential for the biological activity of honey (Brudzynski, 2020; Brudzynski & Sjaarda, 2021).

One of the first studies to characterize the antioxidant properties and polyphenols content of Romania honey was the work of Marghitas et al. (Al et al., 2009). The honeydew had by far the highest polyphenols content, similar to other honey samples from European reports, while the content for floral honey was reported to be lower than the samples reported by other Non-European and European groups. We compared the values of polyphenols content for honey from Romania and other countries and we think that the conclusion was a little bit hasty, as the values reported by Beretta et al., 2005 were both higher and lower than those reported for Romania samples. Indeed, the honey samples from more “exotic” sources such as Malaysian Gelam and Coconut honeys or African samples presented an apparent higher content of polyphenols. We believe that a more reliable comparison could be obtained by analyzing the samples simultaneously and with exactly the same protocol, being known that the Folin-Ciocâlteau method is a very sensitive to interferences and not very specific to polyphenols.

Cimpoiu et al. (2013) performed a detailed investigation on the characteristics of 26 commercial Romanian honeys with different floral origins. Several physical (color intensity, pH, ash content) and biochemical (protein, free amino acid and total phenolic content (TPC), antioxidant activity) were measured and statistically analyzed, and the correlation between TPC and the antioxidant activity was performed. The results showed that Romanian honey has a high antioxidant activity and contains high amounts of valuable compounds. The study also showed that, based on their characteristics, the honeys investigated could be separated in clusters depending on their floral origin. Moreover, high correlation between TPC and the antioxidant activity was obtained, polyphenols contributing with 85.6% to the antioxidant activity.

The results from a more recent study, in which honeydew honey samples from Romania were compared with honey samples from Poland and previous studies confirm that the geographical area and specific climate conditions also influence the phenolic content, beside the origin of honey. The total phenolic and flavonoids content were high and similar between the two countries and highly correlated with the radical scavenging activity (Ma et al., 2016). Oroian et al. (2016) determined for the first time a detailed phenolic profile of honeydew honey samples from the N-E region of Romania (Suceava county) and identified many important polyphenols, with quercetin and pinocembrin in the highest amount (Oroian et al., 2016). Unfortunately, neither the antioxidant activity nor a comparison with other honey samples was performed, so an interpretation from a correlation point of view is not possible. Nevertheless, the high content of polyphenols suggests high antioxidant capacity.

Other parameters responsible for establishing the quality of Romanian honey have been also investigated: composition (total water insoluble matter, diastase and invertase, reducing and easily hydrolysable sugars, volatile organic compounds, and palynological evaluation, as well as the hydroxymethyl furfural (HMF) content, the level of polycyclic aromatic hydrocarbons, and metal levels, adulteration degree, presence of antibiotics and physico-chemical parameters (moisture, electrical conductivity, rheological parameters). These quality parameters were recently reviewed by Isopescu et al., 2017.

Another important biological effect of Romanian honey, the antimicrobial activity, has been also studied in various research papers. It is long known that honey can have significant antimicrobial activity, but its effectiveness was generally shown to depend on honey type and strain specificity. In general, the antibacterial activity is more studied than the antifungal activity of honey, due to the risks raised by the antibiotic resistant bacteria, and this is the case also for the Romanian honey. There is a high demand for new antibacterial formulations to be used in treating or preventing wounds infections. Most Romanian honey types were found to be active against most of the pathogenic bacteria investigated. The most active anti-bacterial honey types were found to be the poly-floral, forest, manna and sunflower honey, while the acacia and linden are generally reported to have moderate antibacterial effects (Fiț et al., 2010; Bobis et al., 2011; Vica et al., 2014; Junie et al., 2016). Some groups reported antibacterial activity against certain strains comparable to the best antibiotics, for some honey types such as forest and poly-floral ones (Fiț et al., 2010). The exact mechanism of the antibacterial activity of honey is not completely known, but there seem to be numerous factors contributing such as enzymatic production of hydrogen peroxide, acidity, pH, high viscosity, low water activity, phytochemicals and other biomolecules, defensin-1, osmotic pressure caused by sugars, etc. A group of Romanian researchers compared several types of honey samples with an artificial honey sample composed of 40.5% fructose, 33.5% glucose, 7.5% maltose, 1.5% sucrose and 17% sterile deionized water. The artificial honey samples, which could be called a natural deep eutectic solvent (NaDES), had no inhibitory effect on any of the eight bacterial species investigated. Contrary to this, all honey samples inhibited to some degree the bacteria strains, the best antibacterial activity capacity was observed for the poly-floral, manna and sunflower and the lowest capacity had the linden and acacia honey. There was a strong correlation between the antibacterial activity and the honey color, the darker honey samples being more effective than the lighter honey

(Junie et al., 2016). The dark color has been shown to be related to pigments and phytochemicals such as carotenoids and flavonoids, based on good correlations between the color and these components (Alvarez-Suarez et al., 2010), which indicates that their contribution to the antibacterial activity is significant.

Table 2. Polyphenols content of Romania honey

Honey type	Region	Pollen type (%)	Polyphenols (mg/100g)
Acacia Lime Sun-flower Honeydew[1]	-	-	TPH (2.00-39.00) TPH (16.00-38.00) TPH (20.00-45.00) TPH (23.00-125.00)
Honeydew[2]	Bihor County	-	TPH (147.0 ± 25.4) FV (15.5 ± 0.5)
Honeydew[3]	N-E Romania (Suceava)	*B. napus* (18.1) *Quercus* (14.3) *T. repens*(11.8) *C. sativa* (8.6) *H. annuus*	pinocembrin (0.3-4.4) quercetin (0.1-2.8) gallic acid (0.02-0.3) galangin (0.02-0.5) myricetin (0-0.4) p-coumaric acid (0-4.4) chrysin (0-0.16) caffeic acid (0-1.9) apigenin (0-1.1) kaempferol (0-0.6) isorhamentin (0-0.12) luteolin (0-0.11)
Acacia Polyflora Lime Forest Sea Buckthorne Sunflower[4]	-	-	TPH (0.76 ± 0.12) TPH (0.78 ± 0.07) TPH (1.04 ± 0.13) TPH (2.38 ± 0.37) TPH (1.33 ± 0.17) TPH (1.17 ± 0.23)

[1](Al et al., 2009)
[2](Ma et al., 2016)
[3](Oroian et al., 2016)
[4](Cimpoiu et al., 2013)

Table 2 summarizes some of the mentioned values for polyphenols content reported in different Romania honey samples. All studies on Romania honey samples indicate that the majority of them conform to the regulation of the European Union, having similar physico-chemical properties to other European honey samples (Isopescu et al., 2014), and that they present high content of valuable bioactive compounds and high antioxidant activities, which mainly correlate with the total content of polyphenols (Cimpoiu et al., 2013).

The antioxidant activity, together with the total phenolic content and individual phenolics, were recently demonstrated to be effective for authentication of Romanian honey (Pauliuc et al., 2020).

The presented data demonstrate that it is possible to improve the perception of the Romanian honey as being a high quality honey, with significant health benefits. Association of the health benefits with the polyphenols content and antimicrobial and/or antioxidant activity could offer a tool similar to UMF, which could further improve the Romanian honey export price.

CONCLUSIONS

Romania is constantly among the top 3 honey producers in European Union. The export price for Romanian honey is rather low, mainly because the traders' efforts are orientated towards counter-acting the structural honey market weakness, such as large number of beekeepers and small apiary size, with a rather low honey yield.

The perception of honey quality by the foreign customers is still not in accordance with the Romanian honey quality. Manuka honey and Mediterranean bitter honey are examples of honey with high price generated by scientific knowledge. Such examples could be followed also for Romanian honey. The content of polyphenols and antioxidant activity could be used not only for the authentication of Romanian honey, but also for substantiation of the beneficial effects on human health.

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