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Article

Phenolic Compounds of Honey Enriched by Wild-Growing Spring Flowers as a New Diet Ingredient

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Abstract: Is it possible that the addition of wild-growing flowers with anti-inflammatory applications can improve the antioxidant properties of rapeseed and multifloral honey? The study objective was a comparative analysis of two honey varieties enriched by flowers of six plant species, based on the content of the antioxidant capacity as well as total flavonoids and phenolic content. Experiments demonstrated that at each level of enrichment there was an improvement in antioxidant properties compared to honey of both varieties without additives, especially in the case of dried flowers. *Primula veris* L. and *Pulmonaria officinalis* L. improved the antioxidant properties and phenolic content most effectively in honey at all levels of enrichment. Rapeseed-type honey produces a better matrix for the incorporation of natural plant metabolites into honey. The content of biologically active substances in honey enriched with flowers gives hope for new applications of health-promoting substances contained in wild plants.

Keywords: anti-inflammatory plant flowers; rapeseed honey; multi-floral honey; antioxidant activity; total phenols; flavonoids

1. Introduction

Honey is an supersaturated aqueous solution of sugar, containing primarily the monosaccharides glucose and fructose. Additionally, it contains a plethora of compounds with various effects, including antioxidants. The health-promoting potential of honey derives specifically from the presence of these antioxidants, both enzymatic and non-enzymatic [1]. The former include catalase and glutathione peroxidase [2]. Non-enzymatic antioxidants include: (i) flavonoids (hesperitin, tricetin, myricetin, pinocembrin, galangin, kaempferol, quercetin, naringenin, chrysin, pinobanksin, luteolin), (ii) non-aromatic organic acids (citric), (iii) phenolic acids and their esters (chlorogenic, gallic, cinnamic, benzoic, caffeic, vanillic, ferulic, abscissic, ellagic), (iv) free amino acids (mainly proline), (v) vitamins (E— α -tocopherol, C—ascorbic acid), (vi) carotenoid derivatives and others [3–5]. Natural honey concentration of phenolic compounds that exhibit antimicrobial and antioxidant properties can exceed 250 $\mu\text{g g}^{-1}$ [6]. Honey has been used since antiquity for its nutritional and medicinal properties. In folk medicine, it is most commonly used as an anti-inflammatory and antibacterial substance, for curing coughing and poorly-healing wounds [7]. Modern medicine also takes advantage of honey's medicinal properties, for example in wound

dressings [8,9], in combating antibiotic-resistant bacteria [10–13], both Gram-positive and Gram-negative [14], for curing coughing [15,16], and in curing acute diarrhea in adults and children [17,18]. A growing demand for honey [19] coupled with honey output being heavily dependent on weather [20], and with the loss of bee families, observed in recent years due to a range of factors [21–23], has been a drive behind the emergence of a new product—herbal honey. Its output is less dependent on weather and can supplement the budget of many professional apiaries. Herbal honey is a honey-like product, produced indirectly by bees fed by the beekeeper with a syrup that contains mainly saccharose, additionally enriched with herbal extracts or fruit juices [4,24]. A comparative study of 3 natural honey varieties and 5 herbal honey types demonstrated that herbal honeys were characterised by a higher antioxidative capacity than natural honeys. Furthermore, the herbal honey with nettle had the highest antibacterial capacity [25]. The antimicrobial effect of herbal honeys against microorganisms, except *E. coli*, is confirmed by a study by Isidorov et al. [26]. However, herbal honeys are generally characterised by a higher saccharose content that permitted for natural honeys [24]. Despite the proven benefits of herbal honeys, from the clinical standpoint it is definitely recommended to use only natural honeys, which are only minimally processed while maintaining their full spectrum of biological activity [27]. Therefore, in order to improve the antioxidative capabilities of beekeeping products, it appears that a better solution is to enrich natural honeys with plant extracts rather than produce herbal honeys. Research demonstrates that the antioxidative activity of honey enriched with mulberry leaves is enhanced even more than 50-fold [28]. An increased phenol content, and consequently improved antioxidative ability, was achieved by enriching natural honey with: algae extracts [29], *Rubus* leaf and flower extracts [30], lavender flowers (*Lavandula* L.), lemon balm flowers (*Melissa* L.), nettle (*Urtica* L.), peppermint flowers (*Mentha* L.) and ginger root (*Zingiber Boehm.*) [31]. By macerating *Sophora* flowers in acacia honey, performed at room temperature for 40 days, a honey enriched in rutin (150.24 mg/kg) and quercetin (1338.93 mg/kg) was produced. For comparison, honey collected from a *Sophora* pasture contained 0.40 mg/kg rutin and 0.10 mg/kg quercetin. (2022). Similarly successful was an attempt to enrich natural honey by macerating *Melilotus officinalis* and *Melilotus albus* flowers (coumarin content increased several-fold) at room temperature for 6 months [32]. Multifloral honeys and natural honeys enriched with plant extracts have very similar physicochemical properties. Conversely, herbal honeys differ in their physicochemical properties, for example having higher pH values than natural and herb-enriched natural honeys [33]. Enriching natural honeys with medicinal plant extracts leads to increased content of pro-health ingredients in the honey. However, only the synergistic effect of honey and herbs, which largely has not been studied, can provide a product effective at curing many ailments. An example is the well-studied synergistic effect of honey and coffee, which has been scientifically verified as a highly effective means of curing persistent post-infection cough [34] and oral cavity mucous membrane inflammation [35].

As the recent years have shown, diseases and infections that society is commonly afflicted with are not only COVID-19, but above all diseases familiar for hundreds of years, such as flu-derived conditions, virus- and bacteria-based conditions covering the upper respiratory tract, as well as RSV viruses that accompany children in particular during early childhood and at the kindergarten age [36]. As a result of the rigorously enforced restrictions and contact limitations, the potential spread of pathogens responsible for children diseases has been effectively contained. As sanitary restrictions were loosened despite the ongoing pandemic and child care and education institutions resumed operations, infections caused by other pathogens rapidly increased, especially in little children. Furthermore, the weakening of immune systems due to various reasons, such as isolation or symptomless Covid-19 infections, resulted in a rising wave of acute influenza and RSV infections, which increasingly necessitated hospitalization of the little patients [37]. At the same time, paediatricians stressed that the only adequate solution is immunity boosting, pro-health prevention practices through food with a high health-promoting potential, and antioxidative, antiviral, and antibacterial properties [38].

Enriching environmentally-friendly honeys with dried plants and extracts from wild-growing plants with high pro-health activity can become a highly effective alternative for food supplements

recommended for immunity boosting against influenza, common cold, and respiratory diseases, especially in children and the youth, who are vulnerable to the RSV. Furthermore, an addition of fragmented dried plants can have a beneficial effect on the condition of the digestive system as a source of dietary fibre that stimulates intestine peristalsis. The presence of pro-health plant ingredients in honey may, for example, reduce its glycemic index, which is an important dietary consideration. In food production flowers, primarily decorative and garden varieties, are used mainly as decorative elements in culinary products [39]. Little robust information is available on the pro-health potential of many wild-growing flowers and flowers of medicinal plants, or the potential has not been thoroughly investigated [40,41]. Literature provides information on the diversity of bioactive compounds found in different parts of medicinal plants, as well as the possibilities for selective extraction of bioactive substances from plant material [42,43]. On the other hand, European Pharmacopoeia [44] states clearly what species and parts of plants are pharmacopoeial medicinal materials and what requirements they must fulfil to be considered as such. Plant-based medicinal product, familiar especially from pharmacotherapy, are characterised by adequate properties that are recognised in medicine and pharmacy, and which are intended to cause a specific therapeutic effect as a result of their application [45]. Their medicinal properties result from specific chemical substances present in those plants in substantial quantities in specific morphological parts, such as roots, leaves, fruits or stones [44]. As a result, it is very frequently that other morphological parts of medicinal plants, while not considered pharmacopoeial materials, contain an equally valuable collection of bioactive compounds, for example, and can become a significant source of said substances in everyday diet provided that the substances are effectively and selectively extracted, that no harmful substances are extracted alongside them, and that they exhibit a sufficiently high biological activity when introduced in a food product, for example as an extract or a dried plant product. Additionally, a potential pharmaceutical material disqualified due to insufficient concentrations of medicinal substances can be a precious and valuable material for food production, providing disease prevention and health boosting properties, although not medicinal qualities, provided that all possible contraindications are analysed. One of the latter can be the presence of plant hormones, saponins or cardiac glycosides, which could potentially be harmful or even dangerous to health [46]. The time of the coronavirus pandemic, time of unceasing restrictions, limitations in contact has brought society closer to nature, or has simply restored the proper respect and importance that nature is due. In recent years numerous studies were undertaken based on traditional medicine, herbal medicine and biomedicine [47].

As a result of the significantly improved nutrition knowledge, consumer behaviors such as greening of consumption, consumer ethnocentrism, self-treatment or subordinating consumer decisions to sustainable lifestyle principles. The 'clean label' idea has gained popularity among consumers, meaning food without additional substances that improve its flavour, shelf life or appearance, very difficult to achieve under the conditions of mass production in the food industry. Biofortifying food with nanocomponents from wild-growing plants, which includes developing technologies for enriching food with functional and health-promoting additives obtained from wild-growing plants, is intended to expand the range of products with a high health-promoting potential and a simple, natural chemical composition. Familiarising consumers with the importance and availability for consumption of wild-growing plants and medicinal plants in other forms than the dried products found in pharmacies and healthy food stores, often not accepted or even completely unknown as Poland has no tradition of consuming herbal infusions, is an important trend in health promotion and disease prevention actions.

In the present study, we investigated the effect of different plants with anti-inflammatory applications on improving honey antioxidant activity. Moreover, the level of total phenols and flavonoids was determined in tested samples. Thus, the results of this work could help understand (1) which plant influences the higher activity of honey and (2) which multifloral or rapeseed honey-type produces the better matrix for plant phenolic incorporation and further their biological activity.

2. Materials and Methods

2.1. Plant material

The plant material in the form of flowers of six species of wild-growing plants (Table 5) from natural ecological habitats located in mountainous areas within the Carpathian Foothills region in south-eastern Poland was gathered at the beginning of April 2022. The test material was cleaned of dust and solids, and dried at 25 °C in a DanLAB (Poland) air circulation laboratory dryer until a water content of 6–8% was achieved. Water content in the dried flower samples was determined using an Ohaus MB12 (Germany) dried scales with an infra-red radiator in accordance with the standard. The dried flowers were ground in an A 11 Basic Analytical Mill (IKA, Germany). The ground plant material was used to enrich honeys and prepare ethanol extracts, which were subsequently used to enrich the honeys as well.

Table 5. Plant species used for honey enrichment.

						
Latin name	<i>Pulmonaria officinalis</i> L. <i>Malva sylvestris</i> L. <i>Primula veris</i> L. <i>Tussilago farfara</i> L. <i>Bellis perennis</i> L. <i>Sambucus nigra</i> L.					
Common name	Lungwort	High mallow	Cowslip primrose	Coltsfoot	Lawndaisy	Black elderberry

Photos Source: <https://commons.wikimedia.org>.

2.2. Preparation of flower extracts

20 g of ground plant material was treated with 100 ml 70% ethanol, the extraction was performed using an ultrasonic bath (Sonic-6D, Polsonic, Warsaw, Poland) at 40 °C for 60 minutes. Once extraction was complete, the samples were moved to a Biosan ES-20/60 (Poland) rotary shaker and the mixture was extracted for 30 minutes at 40 °C and 180 RPM. Once extraction of the mixture was complete, it was filtered under vacuum on Wattman 3 paper filters, and subsequently the extracts were centrifuged at 3 500× g for 20 min. (Eppendorf 5702, Germany). The supernatant was collected, the residue was evaporated under vacuum until dry using a Hei-VAP Precision (Heidolph, Germany) rotary evaporator, then it was dissolved in 50% ethanol and used to enrich the honey samples.

2.3. Honey samples and experimental conditions

Two honey varieties (multifloral and rapeseed) were used for testing, obtained from apiaries located in a mountainous agro-wooded area in the Carpathian Foothills region in south-eastern Poland (49°48'09"N 21°31'53"E). The floral origin of the samples was specified by the beekeepers according to hive location and available floral sources. The honeys were portioned into sterile glass containers with lids, to which plant material in the form of ground dried plants or ethanol extracts

was added and thoroughly, although gently stirred. The plant addition was 1, 2 and 4% (w/w), respectively. For extracts, a volume corresponding to an identical mass of plant material was used. The control samples were honey of both varieties without any plant material added. Honey samples were kept in a dark place at room temperature (21 ± 1 °C) until analysis. The sample curing time was 3 months. During this time, the samples were stirred 3 times. Before further preparation procedures, the honeys were liquefied using an ultrasonic bath (Sonic-6D, Polsonic, Warsaw, Poland) at 40 °C for 1 hour. For testing, 5 g portions from the honey samples were taken into sterile plastic flasks with lids and dissolved in 20 ml water with a 1% acetic acid addition. The aqueous solutions were again placed in an ultrasonic bath for 30 minutes (40 °C), then purified under vacuum on membrane filters (0.45 µm) and evaporated until dry at a Hei-VAP Precision (Heidolph, Germany) vacuum evaporator. The residue was dissolved in 15 ml water with a 1% acetic acid addition. Clear honey solutions were applied in their entirety on conditioned cartridges with a C18 bed (Sep-Pak C18 500 mg, Waters, Ireland). Polyphenolic compounds were eluted with 10 ml methanol directly to a round-bottom flask and evaporated until dry. The residue was dissolved in 2.5 ml MS grade ethanol (Sigma-Aldrich, Poznan, Poland), filtered through nylon filters with 0.22 µm pore diameter (Biospace, Poland), and subsequently analysed.

2.4. Total Antioxidant Capacity

The antioxidant capacity was measured by two different methods: DPPH described by Brand-Williams [48] and ABTS assay based on a procedure reported by Re et al. [49] with slight modifications.

A solution of DPPH (2,2-diphenyl-1-picrylhydrazyl (Sigma-Aldrich, Co. LLC, St. Louis, MO, USA), 0.1 mM in MeOH, was used with an absorbance of 0.95 (± 0.03) at 517 nm. 10 µL of each tested honey sample was added to 140 µL of DPPH solution in wells of a 96-well plate. Absorption was measured after 30 min from the sample addition at 517 nm.

ABTS (2,2'-azino-di-(3-ethylbenzthiazoline sulfonic acid) (Sigma-Aldrich, Co. LLC, St. Louis, MO, USA) was dissolved in water for a 7.5 mM solution. ABTS radical cation (ABTS^{•+}) was generated by reacting 7.5 mM ABTS with 2.5 mM potassium persulfate (final concentration). The mixture was allowed to stand in the dark at room temperature for 16 h before use. The ABTS^{•+} stock solution was diluted with methanol to an absorbance of 0.90 (± 0.02) at 734 nm. The diluted ABTS solution (140 µL) was mixed with 10 µL of the sample prepared from a 50% stock solution of honey sample. The absorption was measured after 30 min from the sample addition at 734 nm. All measurements have been performed using Epoch microplate reader (Biotek Instruments Inc., Winooski, VT, USA).

2.5. Total Phenolic Content (TPC)

Total phenolics were determined by spectrophotometric method using Folin–Ciocalteu reagent (Sigma-Aldrich, Poznan, Poland) described by Jańczak-Pieniążek et al. [50]. Briefly, 75 µL of the sample was mixed with 975 µL of H₂O, then 75 µL of Folin–Ciocalteu reagent (diluted with water 1:1) was added. After 3 min of incubation in darkness at room temperature, 125 µL of 20% Na₂CO₃ (Sigma-Aldrich, Poznan, Poland) was added and mixed. The absorbance of the blue complex was measured at 725 nm using an Epoch microplate spectrophotometer (Biotek Instruments Inc., Winooski, VT, USA).

2.6. Flavonoids Determination

Total flavonoids were determined using the colorimetric method described previously by Zhishen et al. [51]. First, 250 µL of plant extract was mixed with 637 µL of distilled water, then 38 µL of 5% sodium nitrite (Sigma-Aldrich, Poznan, Poland) was added. After 6 min, 75 µL of 10% aluminum chloride (Sigma-Aldrich, Poznan, Poland) solution was added and left to stand for 5 min. Then, 250 µL of 1 M NaOH (Sigma-Aldrich, Poznan, Poland) was added. The absorbance was measured at 510 nm, using an Epoch microplate reader (Biotek Instruments Inc., Winooski, VT, USA). The flavonoid content was expressed as the catechin equivalent.

2.7. Statistical Analysis

All of the analyses were performed in three independent replications for each honey sample. The contents of total phenols, flavonoids, and antioxidant activity were expressed as the mean $\pm$ standard deviation. The obtained results were presented using hierarchical clustering analysis and heatmap visualization. Clustering was performed using the Ward distance matrix that was formed based on the Euclidean distance (data was standardized). Data were analyzed by one-way analysis of variance (ANOVA) using Statistica, v.13.3 (StatSoft, Inc., Tulsa, OK, USA). Significances of differences were calculated using Tukey's multiple range test ($p \leq 0.05$).

3. Results and Discussion

Previously, honey enrichment with wild-growing flowers selected by us was unknown. The flowers came from plants whose therapeutic significance had not always been fully explored before, such as lawndaisy (*Bellis perennis* L.), but the health-promoting properties of other species belonging to the same botanical family were known i.e., Asteraceae [52]. On the other hand, the use of the cowslip (*Primula veris* L.), both in traditional medicine and as a food additive, is well known in South-Eastern European countries [53]. A vast majority of the plant species used in the experiment is known, especially in pharmacy, as plants with medicinal properties and as ingredients of medicines recommended in respiratory diseases (e.g., the Herbapect cough syrup) or as immunity-boosting medicines (e.g., Sinupret), although the therapeutic substances were obtained from morphological parts other than flowers [44,54–56]. Most plants used in our experiments are also identified in literature as potentially effective plants with a range of beneficial qualities, such as antibacterial, anti-inflammatory, antiviral and antioxidative properties [47]. However, they have not been used as food additives, especially as flowers. On the other hand, the choice of honey varieties for the experiment was based on literature reports on their physical (bright colour and stable, semi-liquid consistency), biochemical (relatively low antioxidative potential) and organoleptic (no distinct flavour or odour of their own) properties. Honeys enriched with floral extracts retained their semi-liquid consistency at all levels of enrichment, which for many customers is a quality that improves the value of a product. Sowa et al. [32] used multifloral honey in a study on honey enrichment with *Melilotus* flowers at similar levels as in our study, while Grabek-Lejko et al. [30] enriched rapeseed honey with raspberry fruits and leaves, among others. Based on studies of antioxidative activity of different honey varieties, Dzugan et al. [57] found that it is a certain marker for individual honey varieties. Furthermore they demonstrated that antioxidative activity of rapeseed and multifloral honeys was the lowest, and that they were distinguished by the brightest colour from among the test honey varieties, which supports the selection of these two varieties for our experiment.

The two most commonly used tests, DPPH and ABTS, were used to assess the anti-oxidative properties of honeys enriched with flowers and floral extracts in our study. The antioxidative tests are in vitro methods designed to imitate the oxidation and reduction reactions occurring in living biological systems to assess the antioxidative potential of different chemical and biological samples. According to Gil et al. [58], the ABTS test values were in general significantly higher than the DPPH test. Nevertheless, they should be considered a confirmation of the DPPH test. This is also confirmed by Aebisher et al. [59], who in their study on determinations of the antioxidative activity of essential oils demonstrated that the synthetic antioxidants butylated hydroxyanisole and butylated hydroxytoluene, which contain a phenol ring, strongly capture ABTS in comparison with the DPPH radical. The results of our total Antioxidant Capacity determinations in both tests are presented as percentage values to show how effectively the synthetic radical is neutralised in honeys with plant additives, because it is increasingly frequently suggested that synthetic antioxidants, such as BHA and BHT, exhibit toxic properties and are potentially harmful to human health [60,61]. Table 1 shows the average antiradical activity results for honeys enriched with flowers, determined using the DPPH method.

Table 1. Total antioxidant activity (expressed in percentage $\pm$ standard deviation) of tested honey products against synthetic the DPPH radical.

Sample/Plant Species	Form of enrichment	Concentration		
		1%	2%	4%
Rapeseed honey	No plant addition		1.61 $\pm$ 0.93 u	
<i>Pulmonaria officinalis</i> L.	Extract	5.83 $\pm$ 2.74 stu	11.43 $\pm$ 2.14 oprst	19.64 $\pm$ 1.79 klmn
	Dried flowers	24.84 $\pm$ 1.59 jkl	38.61 $\pm$ 5.26 gh	67.10 $\pm$ 1.79 bc
<i>Malva sylvestris</i> L.	Extract	5.16 $\pm$ 0.16 tu	8.29 $\pm$ 0.36 rstu	15.28 $\pm$ 1.31 mnopr
	Dried flowers	9.64 $\pm$ 1.31 prstu	17.09 $\pm$ 2.38 lmnop	73.61 $\pm$ 3.64 b
<i>Primula veris</i> L.	Extract	13.89 $\pm$ 0.95 noprs	18.17 $\pm$ 1.75 klmno	41.43 $\pm$ 0.08 gh
	Dried flowers	25.79 $\pm$ 5.24 jk	72.66 $\pm$ 4.96 b	83.06 $\pm$ 0.99 a
<i>Tussilago farfara</i> L.	Extract	15.44 $\pm$ 0.04 mnopr	29.60 $\pm$ 1.43 ij	41.86 $\pm$ 2.34 fgh
	Dried flowers	23.13 $\pm$ 2.82 jklm	44.32 $\pm$ 0.36 efg	58.73 $\pm$ 5.15 cd
<i>Bellis perennis</i> L.	Extract	7.74 $\pm$ 2.98 rstu	13.06 $\pm$ 1.39 noprs	22.88 $\pm$ 2.40 jklm
	Dried flowers	14.90 $\pm$ 2.18 mnopr	25.66 $\pm$ 4.03 jk	42.14 $\pm$ 4.05 fgh
<i>Sambucus nigra</i> L.	Extract	20.28 $\pm$ 0.59 klmn	35.30 $\pm$ 2.56 hi	51.31 $\pm$ 3.21 de
	Dried flowers	21.27 $\pm$ 0.24 jklmn	50.24 $\pm$ 0.87 ef	66.27 $\pm$ 0.63 bc
Multifloral honey	No plant addition		3.76 $\pm$ 0.58 s	
<i>Pulmonaria officinalis</i> L.	Extract	14.11 $\pm$ 0.87 nopr	20.48 $\pm$ 0.48 lmn	34.23 $\pm$ 2.81 gh
	Dried flowers	28.84 $\pm$ 2.39 hij	43.84 $\pm$ 2.29 de	80.32 $\pm$ 0.58 a
<i>Malva sylvestris</i> L.	Extract	8.87 $\pm$ 1.32 prs	14.64 $\pm$ 1.81 nopr	17.58 $\pm$ 0.35 lmn
	Dried flowers	14.58 $\pm$ 0.84 nopr	28.39 $\pm$ 2.58 ijk	35.97 $\pm$ 1.58 fg
<i>Primula veris</i> L.	Extract	8.35 $\pm$ 1.32 rs	14.16 $\pm$ 0.29 nopr	28.90 $\pm$ 0.58 hij
	Dried flowers	50.29 $\pm$ 2.81 c	74.64 $\pm$ 1.11 a	80.26 $\pm$ 3.16 a
<i>Tussilago farfara</i> L.	Extract	13.06 $\pm$ 0.74 opr	23.23 $\pm$ 0.77 jkl	49.81 $\pm$ 4.84 cd
	Dried flowers	14.71 $\pm$ 0.90 nopr	22.35 $\pm$ 2.30 klm	48.28 $\pm$ 4.76 cd
<i>Bellis perennis</i> L.	Extract	10.87 $\pm$ 1.00 opr	9.35 $\pm$ 0.34 prs	23.74 $\pm$ 0.32 jkl
	Dried flowers	16.29 $\pm$ 0.68 mno	26.93 $\pm$ 0.93 ijk	35.39 $\pm$ 1.97 fg
<i>Sambucus nigra</i> L.	Extract	14.90 $\pm$ 2.13 nop	29.61 $\pm$ 2.32 ghij	41.32 $\pm$ 1.58 ef
	Dried flowers	32.45 $\pm$ 1.61 ghi	47.00 $\pm$ 3.00 cde	63.42 $\pm$ 0.19 b

*—Results marked by different letters are statistically different at $P \leq 0.05$ Tuckey's test.

The highest synthetic radical neutralisation capacity was observed in rapeseed honeys with a 4% dried cowslip flower addition, and it was more than 50 times higher than the activity for pure rapeseed honey. A similar effect was observed for enrichment with *Malva sylvestris* L. flowers added in similar quantities as *Primula veris* L., and *Primula veris* L. flowers in multifloral honeys with a 4% dried flowers addition. Rapeseed honeys with an addition of *Pulmonaria officinalis* L. and *Sambucus nigra* L. flowers and multifloral honeys enriched with *Sambucus nigra* L. flowers were characterised by an antioxidative activity of 65.60% on average, lower by approx. 20% compared with the highest results in this group, observed for the 4% plant addition. The lowest values for the 4% plant addition were noted for rapeseed honeys with added lungwort and high mallow, and multifloral honeys with common daisy and high mallow extract, ranging from 23.74 to 15.28%. For the 2% plant addition, the highest antioxidative activity was observed for multifloral honeys with *Primula veris* L. flowers. The second highest were multifloral and rapeseed honeys with *Sambucus nigra* L. flowers, and rapeseed honeys with *Tussilago farfara* L. flowers, as well as multifloral honeys with *Pulmonaria officinalis* L. flowers, whose antioxidative activity was within the 50.24 to 43.84% range. Unlike multifloral honeys enriched with dried cowslip, the same products enriched with extracts from these flowers exhibited the lowest antioxidative properties (14.16%) in the 2% enrichment group, similarly to honeys with daisy extract (9.35%) and rapeseed honeys with lungwort and high mallow extract, 11.43% and 8.29%, respectively. Enriching honeys at the 1% plant addition level confirmed the highest antioxidative activity of multifloral honeys enriched with the *Primula veris* L. extract, which was 50.29%. Honeys enriched with elderberry flowers exhibited approximately 40% lower antioxidative activity than honeys with cowslip. Among the rapeseed honeys enriched with 1% flowers, honeys with dried cowslip, lungwort and coltsfoot exhibited the highest antiradical activity. The lowest antioxidative

activity was observed for multifloral and rapeseed honeys enriched with *Malva sylvestris* L., *Primula veris* L. and *Bellis perennis* L. (multifloral honeys) and *Pulmonaria officinalis* L. (rapeseed honeys) floral extracts. As a result of enriching honeys with flowers and extracts from wild-growing spring flowers, the antioxidative properties were improved in every case, compared to honeys of the two varieties without the additions, but much higher results were observed for dried flower additions than for flower extracts. Among all the flower additions tested, dried cowslip was the most effective at improving the antioxidative properties of honey determined using the DPPH method at all enrichment ratios, with a high effectiveness of the extract was observed for the 2% addition, where the antioxidative activity observed was almost 3 times higher than for a flower addition smaller by half, and only 7% lower than for a dried cowslip addition twice as large.

For the antioxidative activity test by the ABTS method (Table 2), higher values were observed than with the DPPH test both for honey samples without additions and for enriched honeys at all levels, which confirms the previously mentioned report by Gil et al. [58].

Table 2. Total antioxidant activity (expressed in percentage $\pm$ standard deviation) of tested honey products against synthetic the ABTS radical.

Sample/Plant Species	Form of enrichment	Concentration		
		1%	2%	4%
Rapeseed honey	No plant addition		15.98 $\pm$ 0.31 s	
<i>Pulmonaria officinalis</i> L.	Extract	25.00 $\pm$ 1.82 pr	43.29 $\pm$ 1.18 klm	58.04 $\pm$ 4.78 hi
	Dried flowers	73.13 $\pm$ 3.20 ef	99.51 $\pm$ 0.11 a	99.85 $\pm$ 0.05 a
<i>Malva sylvestris</i> L.	Extract	21.35 $\pm$ 0.94 rs	40.29 $\pm$ 0.15 lmn	70.91 $\pm$ 0.69 fg
	Dried flowers	48.37 $\pm$ 2.71 jk	69.58 $\pm$ 0.34 fg	99.90 $\pm$ 0.00 a
<i>Primula veris</i> L.	Extract	38.21 $\pm$ 3.89 mno	54.59 $\pm$ 3.81 ij	92.06 $\pm$ 0.84 bc
	Dried flowers	84.86 $\pm$ 0.34 d	99.90 $\pm$ 0.00 a	100.00 $\pm$ 0.00 a
<i>Tussilago farfara</i> L.	Extract	31.51 $\pm$ 0.05 op	55.32 $\pm$ 0.89 ij	86.59 $\pm$ 4.44 cd
	Dried flowers	64.30 $\pm$ 4.34 gh	98.18 $\pm$ 0.25 ab	99.70 $\pm$ 0.11 a
<i>Bellis perennis</i> L.	Extract	20.81 $\pm$ 0.31 rs	34.21 $\pm$ 0.94 no	70.22 $\pm$ 1.46 fg
	Dried flowers	46.10 $\pm$ 2.42 kl	70.76 $\pm$ 1.63 fg	79.49 $\pm$ 1.48 de
<i>Sambucus nigra</i> L.	Extract	54.59 $\pm$ 3.01 ij	70.96 $\pm$ 4.98 fg	94.67 $\pm$ 2.17 ab
	Dried flowers	48.37 $\pm$ 2.71 gh	69.58 $\pm$ 0.34 ab	99.91 $\pm$ 0.00 a
Multifloral honey	No plant addition		13.49 $\pm$ 0.59 o	
<i>Pulmonaria officinalis</i> L.	Extract	30.73 $\pm$ 1.16 mn	55.68 $\pm$ 1.64 gh	90.08 $\pm$ 6.84 bcd
	Dried flowers	88.25 $\pm$ 0.00 cd	99.61 $\pm$ 0.00 a	99.85 $\pm$ 0.14 a
<i>Malva sylvestris</i> L.	Extract	28.52 $\pm$ 1.16 n	44.91 $\pm$ 3.08 ijkl	56.33 $\pm$ 2.31 gh
	Dried flowers	52.54 $\pm$ 2.21 ghi	80.15 $\pm$ 1.86 de	93.79 $\pm$ 1.54 abc
<i>Primula veris</i> L.	Extract	41.18 $\pm$ 3.23 kl	58.62 $\pm$ 9.78 fg	78.13 $\pm$ 3.66 e
	Dried flowers	97.41 $\pm$ 0.87 abc	99.85 $\pm$ 0.05 a	99.85 $\pm$ 0.05 a
<i>Tussilago farfara</i> L.	Extract	38.44 $\pm$ 2.71 lm	59.63 $\pm$ 5.20 fg	98.84 $\pm$ 0.19 ab
	Dried flowers	44.03 $\pm$ 0.58 ijk	54.13 $\pm$ 2.61 ghi	98.84 $\pm$ 0.38 ab
<i>Bellis perennis</i> L.	Extract	28.76 $\pm$ 0.43 n	41.57 $\pm$ 4.29 jkl	66.86 $\pm$ 1.06 f
	Dried flowers	50.77 $\pm$ 0.38 ghij	66.28 $\pm$ 2.50 f	83.38 $\pm$ 3.99 de
<i>Sambucus nigra</i> L.	Extract	48.41 $\pm$ 2.84 hijk	66.52 $\pm$ 1.20 f	98.12 $\pm$ 0.72 ab
	Dried flowers	79.38 $\pm$ 1.93 de	98.89 $\pm$ 0.05 ab	99.52 $\pm$ 0.10 a

*—Results marked by different letters are statistically different at $P \leq 0.05$ Tuckey's test.

A 30-minute incubation of the extracts resulted in almost 100% neutralisation of the synthetic radical for rapeseed and multifloral honeys with cowslip, elderberry, high mallow, lungwort and coltsfoot additions, especially for 4% dried flower addition, while for multifloral honeys, a similar effect of almost 100% was observed for honeys enriched with cowslip flowers. On the other hand, multifloral honey without additions exhibited an antioxidative activity of 13.49%, which was lower by approximately 15.0% than that of rapeseed honey without additions. The lowest antioxidative potential of 20.81% characterised rapeseed honey samples with 1% *Bellis perennis* L. flower extract, which was higher by 23% than the antioxidative potential value of this honey variety without an

addition, while for a 4 times greater extract or dried flowers addition, the antioxidative activity values observed were higher by 70% and 80%, respectively, than in samples without additions, both for rapeseed and multifloral honey. The best improved antioxidative potential was observed in rapeseed and multifloral honeys when enriched with dried *Primula veris* L. at all addition ratios, with even 1% addition in multifloral honeys providing almost complete neutralisation of the synthetic radical (97.40%). In the same honeys with the lowest enrichment ratio, a high result of 88.23% was observed with the addition of *Pulmonaria officinalis* L. and *Sambucus nigra* L. flowers (79.38%), which was almost 6 times higher than pure (no additions) multifloral honey. The lowest antiradical activities characterised honey samples of both varieties at all enrichment ratios when extracts of *Bellis perennis* L., *Malva sylvestris* L. and *Pulmonaria officinalis* L. (only rapeseed honeys) were added at three enrichment ratios. The second stage of the experiment consisted of the analysis of total phenolic content (TPC) in honeys enriched with dried flowers and floral extracts. The test results are presented in Table 3.

Table 3. The content of total phenols (mg of gallic acid equivalent per 100 g) in tested honey products.

Sample/Plant Species	Form of enrichment	Concentration		
		1%	2%	4%
Rapeseed honey	No plant addition		2.24 ± 0.01 s	
<i>Pulmonaria officinalis</i> L.	Extract	4.72 ± 0.21 nop	6.80 ± 0.24 jkl	9.81 ± 0.32 fgh
	Dried flowers	8.22 ± 0.16 i	9.84 ± 0.31 gh	12.23 ± 0.61 cd
<i>Malva sylvestris</i> L.	Extract	3.55 ± 0.37 r	5.12 ± 0.30 mno	6.10 ± 0.15 klm
	Dried flowers	4.70 ± 0.44 nop	7.81 ± 0.45 ij	9.16 ± 0.44 hi
<i>Primula veris</i> L.	Extract	6.17 ± 0.43 klm	8.76 ± 0.40 hi	10.84 ± 0.32 efg
	Dried flowers	1.15 ± 0.45 de	13.44 ± 0.63 ab	13.65 ± 0.32 a
<i>Tussilago farfara</i> L.	Extract	5.83 ± 0.4 l mn	6.40 ± 0.16 kl	11.00 ± 0.20 ef
	Dried flowers	4.55 ± 0.26 opr	8.78 ± 0.12 hi	12.42 ± 0.63 bc
<i>Bellis perennis</i> L.	Extract	3.71 ± 0.34 pr	5.23 ± 0.27 mno	7.01 ± 0.33 jk
	Dried flowers	7.98 ± 0.22 ij	9.51 ± 0.20 h	11.44 ± 0.55 cde
<i>Sambucus nigra</i> L.	Extract	8.34 ± 0.20 i	10.4 ± 0.52 efg	12.18 ± 0.52 cd
	Dried flowers	8.83 ± 0.10 hi	12.2 ± 0.60 bcd	14.16 ± 0.41 a
Multifloral honey	No plant addition		3.56 ± 0.15 n	
<i>Pulmonaria officinalis</i> L.	Extract	3.76 ± 0.50 n	6.52 ± 0.10 kl	9.77 ± 0.58 gh
	Dried flowers	5.71 ± 0.18 lm	12.63 ± 0.31 cd	13.03 ± 0.54 cd
<i>Malva sylvestris</i> L.	Extract	5.32 ± 0.32 m	8.54 ± 0.00 ij	10.90 ± 0.33 efg
	Dried flowers	5.58 ± 0.34 m	8.12 ± 0.40 j	13.68 ± 0.15 ab
<i>Primula veris</i> L.	Extract	5.88 ± 0.11 lm	9.50 ± 0.33 hi	13.53 ± 0.20 abc
	Dried flowers	7.50 ± 0.39 jk	12.01 ± 0.26 de	14.21 ± 0.13 ab
<i>Tussilago farfara</i> L.	Extract	5.42 ± 0.62 m	6.87 ± 0.77 kl	10.00 ± 0.35 gh
	Dried flowers	8.37 ± 0.36 ij	10.81 ± 0.42 fg	13.33 ± 0.16 bc
<i>Bellis perennis</i> L.	Extract	3.69 ± 0.24 n	6.58 ± 0.39 kl	12.07 ± 0.13 de
	Dried flowers	6.10 ± 0.66 lm	10.50 ± 0.21 fgh	13.13 ± 0.34 bcd
<i>Sambucus nigra</i> L.	Extract	6.55 ± 0.32 kl	10.44 ± 0.36 fgh	13.40 ± 0.12 bc
	Dried flowers	11.30 ± 0.10 ef	14.30 ± 0.13 a	14.61 ± 0.26 a

*—Results marked by different letters are statistically different at $P \leq 0.05$ Tuckey's test.

The highest total phenolic content characterised multifloral honeys enriched with elderberry and cowslip at 4% plant material addition, both as an extract and as dried flowers, and honey enriched with high mallow, and exceeded the total phenolic content in multifloral honey samples, which was 3.56 mg GAE/100 g⁻¹, almost 4 times. A similar phenolic content as in samples with a 4% dried flowers addition, was observed for a 2% addition of the *Sambucus nigra* L., *Pulmonaria officinalis* L. and *Primula veris* L. flowers listed previously. Therefore, this ratio can be considered optimal enrichment for these plant species, as higher content does not significantly affect the phenolic content. On the other hand, for a 1% plant material addition, the highest total phenolic content was observed for enrichment with elderberry flowers, the value being 11.30 mg GAE/100 g⁻¹. The dried elderberry flower addition

caused a TPC increase at each of the 3 plant matter addition ratios in multifloral honeys. The lowest total phenolic content was noted for *Pulmonaria officinalis* L. extract addition at each enrichment ratio in multifloral honeys, with TPC for 1% addition not differing significantly from the phenolic content in the honey sample without plant material added. For rapeseed honeys enriched with a 4% plant material addition, the total phenolic content was observed for honeys enriched with *Sambucus nigra* L., *Primula veris* L., *Tussilago farfara* L. and *Pulmonaria officinalis* L., the values ranging from 14.16 mg GAE·100 g⁻¹ (elderberry) to 12.23 mg GAE·100 g⁻¹ (lungwort). These values were 6 times higher than in honey samples without additions. Conversely, the lowest TPC in the 4% plant addition group in rapeseed honey was found for daisy extract, 7.01 mg GAE·100 g⁻¹, and for high mallow at 6.10 mg GAE·100 g⁻¹. In rapeseed honeys, a dried cowslip flower addition of just 2% resulted in an equally high total phenolic content, 13.44 mg GAE·100 g⁻¹ on average, as with a dried flower addition twice as large, while honey with an extract addition at the same ratio showed a 35% lower total phenolic content than rapeseed honey enriched with dried *Primula veris* L. flowers, for a value of 8.76 mg GAE·100 g⁻¹. Rapeseed honey enrichment with cowslip flowers significantly increased the total phenolic content in the honey samples at all addition ratios compared to honeys without additions, with the greatest increase noted at 1% and 2% additions. The greatest increase in total phenolic content was observed for cowslip flowers, which was lower by 17% than the twice-higher enrichment in rapeseed honeys. As with multifloral honeys, the lowest phenolic content was observed in rapeseed honeys with the lowest plant addition, in particular samples enriched with *Malva sylvestris* L. (3.55 mg GAE·100 g⁻¹) and *Bellis perennis* L. extracts (3.71 mg GAE·100 g⁻¹). Table 4 shows the test results of flavonoid content in rapeseed and multifloral honeys enriched with flowers.

Table 4. The content of flavonoids (mg of quercetin equivalent per 100 g) in tested honey products.

Sample/Plant Species	Form of enrichment	Concentration		
		1%	2%	4%
Rapeseed honey	No plant addition		8.42 ± 0.66 mn	
<i>Pulmonaria officinalis</i> L.	Extract	6.72 ± 0.21 no	16.12 ± 0.90 j	28.26 ± 0.63 e
	Dried flowers	16.24 ± 0.56 j	33.31 ± 0.67 d	61.00 ± 0.86 a
<i>Malva sylvestris</i> L.	Extract	5.85 ± 0.52 o	10.64 ± 0.23 lm	14.36 ± 0.68 jk
	Dried flowers	9.62 ± 0.33 lm	18.37 ± 0.75 i	24.04 ± 0.92 g
<i>Primula veris</i> L.	Extract	8.91 ± 0.14 m	14.55 ± 0.92 jk	24.71 ± 0.54 g
	Dried flowers	14.65 ± 0.83 jk	26.51 ± 0.80 ef	33.62 ± 0.72 d
<i>Tussilago farfara</i> L.	Extract	11.34 ± 0.66 lm	24.82 ± 0.09 fg	37.55 ± 0.99 c
	Dried flowers	9.36 ± 0.51 lm	20.65 ± 0.63 h	40.13 ± 0.88 b
<i>Bellis perennis</i> L.	Extract	10.30 ± 0.24 lm	15.26 ± 0.32 jk	23.18 ± 0.67 g
	Dried flowers	9.26 ± 0.21 m	14.92 ± 0.36 jk	23.94 ± 0.38 g
<i>Sambucus nigra</i> L.	Extract	13.64 ± 0.65 k	24.60 ± 0.29 fg	37.43 ± 0.43 c
	Dried flowers	27.92 ± 0.82 e	33.82 ± 0.61 d	39.36 ± 0.42 bc
Multifloral honey	No plant addition		4.33 ± 0.30 t	
<i>Pulmonaria officinalis</i> L.	Extract	9.03 ± 0.51 r	21.12 ± 0.76 gh	22.82 ± 0.77 fg
	Dried flowers	15.35 ± 0.60 lm	29.87 ± 0.58 d	40.87 ± 0.78 a
<i>Malva sylvestris</i> L.	Extract	4.32 ± 0.14 t	9.10 ± 0.62 r	16.86 ± 0.86 kl
	Dried flowers	9.27 ± 0.33 r	11.45 ± 0.56 p	35.53 ± 0.61 b
<i>Primula veris</i> L.	Extract	4.03 ± 0.45 t	9.42 ± 0.31 r	13.87 ± 0.47 mn
	Dried flowers	14.64 ± 0.56 mn	18.80 ± 0.71 ij	24.38 ± 0.52 f
<i>Tussilago farfara</i> L.	Extract	5.05 ± 0.53 st	11.77 ± 0.34 op	26.84 ± 0.67 e
	Dried flowers	13.05 ± 0.34 no	20.65 ± 0.41 hi	27.26 ± 0.43 e
<i>Bellis perennis</i> L.	Extract	6.32 ± 0.28 s	9.20 ± 0.30 r	22.44 ± 0.38 gh
	Dried flowers	12.51 ± 0.55 op	20.67 ± 0.68 hi	30.82 ± 0.65 d
<i>Sambucus nigra</i> L.	Extract	11.62 ± 0.68 op	18.92 ± 0.66 ij	36.90 ± 0.57 b
	Dried flowers	17.44 ± 0.42 jk	32.90 ± 0.39 c	40.01 ± 0.89 a

*—Results marked by different letters are statistically different at $P \leq 0.05$ Tuckey's test.

The highest flavonoid content was noted in rapeseed honeys enriched with 4% dried lungwort, with the average value of 61.00 mg QE·100 g⁻¹ being more than 7 times higher than for pure rapeseed honeys. In the same products enriched with a floral extract, a flavonoid content lower by half was found. On the other hand, in honeys enriched with coltsfoot and elderberry flowers, flavonoid content was almost 5 times higher, compared to honey samples without additions, both for honeys enriched with floral extracts and with dried flowers. A 4% common daisy addition significantly increased flavonoid content in honeys, almost 3 times higher than for rapeseed honeys without plant additions, where the content was 4.33 mg QE·100 g⁻¹. The lowest flavonoid content, despite a 4% enrichment, was noted for honeys with added *Primula veris* L. and *Malva sylvestris* L. extracts, where a flavonoid content increase by 70% and 185%, respectively, was noted in comparison with honey samples without additions. Rapeseed honey enrichment with dried lungwort, elderberry, and cowslip flowers at all addition ratios resulted in the greatest increases in flavonoid content within their respective groups. For honeys with coltsfoot and daisy, a similar effectiveness of plant material enrichment was observed for extracts and dried flowers. Multifloral honeys were characterised by a lower flavonoid content than rapeseed honeys, but produced higher results after plant material was added. The highest enrichment effectiveness was observed for dried lungwort, elderberry and high mellow additions, where flavonoid content increased more than 9 times compared to honey samples without additions. The additions of lungwort and elderberry flowers were the most effective for increasing flavonoid content at all enrichment ratios. Statistically highly significant differences were found in flavonoid content for all levels of enrichment and for each plant species, similar as with the preceding determinations.

According to different authors, Polish rapeseed honey is relatively poor in phenolic compounds, containing from approx. 4.5 to approx. 33.5 mg GAE·100 g⁻¹ [62]. A study conducted by Džugan et al. [57] demonstrated that rapeseed honeys had an antioxidative activity only half that of multifloral honeys, which is consistent with our results. Wilczyńska et al. [63] reported that the antiradical activity measured by the DPPH method ranged from 23.8% in Polish nectar-honeydew honeys to 100% for Polish heather and buckwheat honeys. Jasicka-Misiak et al. [64] reported similar values (31-40%) for Polish woundwort honey, measured for a 20% w/v honey solution. In turn, Lachman et al. [65] found, after analysing multiple varieties of Czech honeys, antioxidant activity determined by the DPPH, ABTS methods was lowest in floral honeys. Bertoncelj et al. [66] observed a low antioxidative activity in Slovenian honeys with the brightest colour, which were the acacia and linden varieties, while the lowest values were found for dark honeys: fir, spruce, and forest honey. Perna et al. [67] tested Italian honeys and discovered that radical sweeping activity measured for honey solutions of 3-60% w/v concentration ranged from 55.06% for citrus honey to 75.37% for chestnut honey. Investigating the antioxidative activity of Serbian honeys, Srećković et al. [68] found that forest honey showed better antioxidative activity, on average 594.77 mg Trolox·kg⁻¹ in the ABTS test, and 260.77 mg Trolox·kg⁻¹ in the DPPH test, than other test samples of honey. In turn, a determination of phenolic content by the same authors using the spectrophotometric method in honey samples demonstrated that the highest total phenolic content (806.10 mg GAE·kg⁻¹) and flavonoid content (146.27 mg Qu·kg⁻¹) was found for forest honey and was more than ten times higher than for acacia honey, in which total phenolic content was determined at 68.48 mg GAE·kg⁻¹, and flavonoid content at 18.59 mg QU·kg⁻¹. According to Kacániová et al. [69] the radical scavenging activity in Slovakian honeydew honey samples measured for a 25% w/v honey solution ranged from 45.9 to 86.6%. Concerning the content of phenolic compounds determined as total phenolic content, especially for multifloral honeys, the authors of many studies report highly varied data. For example, Kavanagh et al. [70] found that in multifloral Irish honeys, total phenolic content (TPC) ranged from 2.59 to 81.10 mg GAE·100 g⁻¹ honey. In the authors' opinion, the result of this determination was affected by the region where the honey was collected. In Irish honeys produced in rural areas, a much lower total phenolic content was recorded (20.32 mg GAE·100 g⁻¹) than for city honeys from the same region (28.26 mg GAE·100 g⁻¹) due to a much greater diversity of floral resources in Irish urban areas. Muñoz et al. [71] reported that Peruvian wild multifloral honey showed the highest phenolic compound content, which was 207.89 ± 2.18 mg GAE·100 g⁻¹, with the results also showing the lowest variability.

It also bears highlighting that there are monofloral honeys characterised by a high phenolic compound content, which leads to a high interest in these products, for example the Manuka honey (*Leptospermum scoparium*), which originates from New Zealand or Australia, is distinguished by the highest total phenolic content recorded to date, ranging from 217.0 to 203.0 mg GAE $\cdot 100\text{ g}^{-1}$, as well as a high antioxidative and antibacterial activity, and is considered a medicinal honey. The Polish equivalent to this product, according to Golinski et al. [72], is buckwheat honey, whose TPC value is 211.0 ± 11.4 mg GAE $\cdot 100\text{ g}^{-1}$ and does not differ significantly from that of Manuka honey. Becerril-Sánchez et al. [73], based on an extensive review of previous studies on antioxidative properties and phenolic and flavonoid content in honey, pointed to these parameters as the characteristics that distinguished individual varieties, botanical origin, or even allowed honey falsification to be detected and indicate its health promoting applications. The transformations of flavonoid compounds affect not only the biochemistry and physiology of the plants, acting as antioxidants, enzyme inhibitors, but also as substances with beneficial properties for humans, affecting certain aspects of metabolism; therefore, their presence in daily diet is extremely important [74]. For pure raw honeys, flavonoid content differs between varieties, ranging from 0.56–0.62 mg QE $\cdot 100\text{ g}^{-1}$ for multifloral and rape honeys to 0.53–0.90 mg QE $\cdot 100\text{ g}^{-1}$ for forest honey [65].

Introducing a plant ingredient, both during the honey production stage as well as through enrichment in the form of an extract or fragmented plant material, can effectively increase flavonoid content in honey. A high flavonoid content distinguished the herb honeys tested by Socha et al. [5], and especially thyme herb honey, hawthorn herb honey and raspberry herb honey, whose total flavonoids ranged from 20 to 28 mg QE $\cdot 100\text{ g}^{-1}$, and additionally for these three products, the investigators also noted a very high antioxidative activity. Conversely, in the study conducted by Grabek-Lejko et al. [30], the highest flavonoid content characterised rapeseed honeys enriched with blackberry and raspberry leaves, with the honeys also exhibiting a high antioxidative activity. Tomczyk et al. [28], who enriched rapeseed honeys with mulberry leaves, found a 50-fold increase in antioxidative potential. Additionally, they found that the high antioxidative potential of mulberry-enriched honeys resulted primarily from the presence of phenolic acids and flavonoid glycosides. Jović et al. [47] demonstrated in their study a high pro-health potential of nineteen extracts from the leaves and flowers of medicinal plants and herbs, including cowslip flower, elderberry flower, and high mallow extracts. Among these, the cowslip extract was characterised by the highest antioxidative potential of 188.5 GAE mg $\cdot\text{g}^{-1}$, elderberry extract placing second with an antioxidative activity of 170.4 GAE mg $\cdot\text{g}^{-1}$, while high mallow had a potential 10 times lower than elderberry flowers. Flavonoid content was 52.0 RUE mg $\cdot\text{g}^{-1}$ (*Primula veris* L.), 32.4 RUE mg $\cdot\text{g}^{-1}$ (*Sambucus nigra* L.) and 35.5 RUE mg $\cdot\text{g}^{-1}$ (*Malva sylvestris* L.), respectively. These test results demonstrate that individual flowers and other plant parts can provide different health promoting properties and substances, while antioxidative activity is not always connected to a high flavonoid content. A study conducted by Tarapatskyy et al. [43] demonstrated that the richest source of polyphenolic compounds was cowslip primrose flowers and leaves, while aqueous and ethanol extracts from *Primula veris* L. were characterised by a quantitatively rich profile of polyphenolic substances and a high antioxidative potential. According to Wichtl [75], total flavonoid content in cowslip flowers reaches approximately 3%, and the substances present in these flowers in the greatest quantities are rutoside, kaempferol-3-rutinoside, and isorhamnetin-3-glucoside. Tarapatskyy et al. [42] additionally demonstrate that as a result of enriching wine with cowslip flowers, anthocyanin content in red wine increased 4 times up to 1956.85 mg $\cdot\text{L}^{-1}$ after a 10% addition of *Primula veris* L. flowers, flavonol content increased 5 times for white wines, and an almost 25-fold increase in flavonol content was found in Carlo Rossi commercial wine samples at the lowest (2.5%) *Primula veris* L. flower addition. In a Latypova et al. [76] study, the object of analysis was a solid herbal extract of *Primula veris* L., which included a multi-stage purification process with standardisation of its polyphenolic composition. The therapeutic effect of the extract on the myocardial contractile function in animals with experimental chronic heart failure (CHF) was then examined. The authors of the study demonstrated that a solid herbal extract produced from *Primula veris* L. at a dose of 30 mg $\cdot\text{kg}^{-1}$ exerts a cardioprotective effect

which is evidenced by a smaller number of animal deaths, a lower level of CHF plasma markers, and a greater increase in myocardial contraction and relaxation rates as compared to the control group.

Hierarchical clustering analysis and heatmap visualization, was performed to visualize the relationships between the analyzed samples (Figure 1).

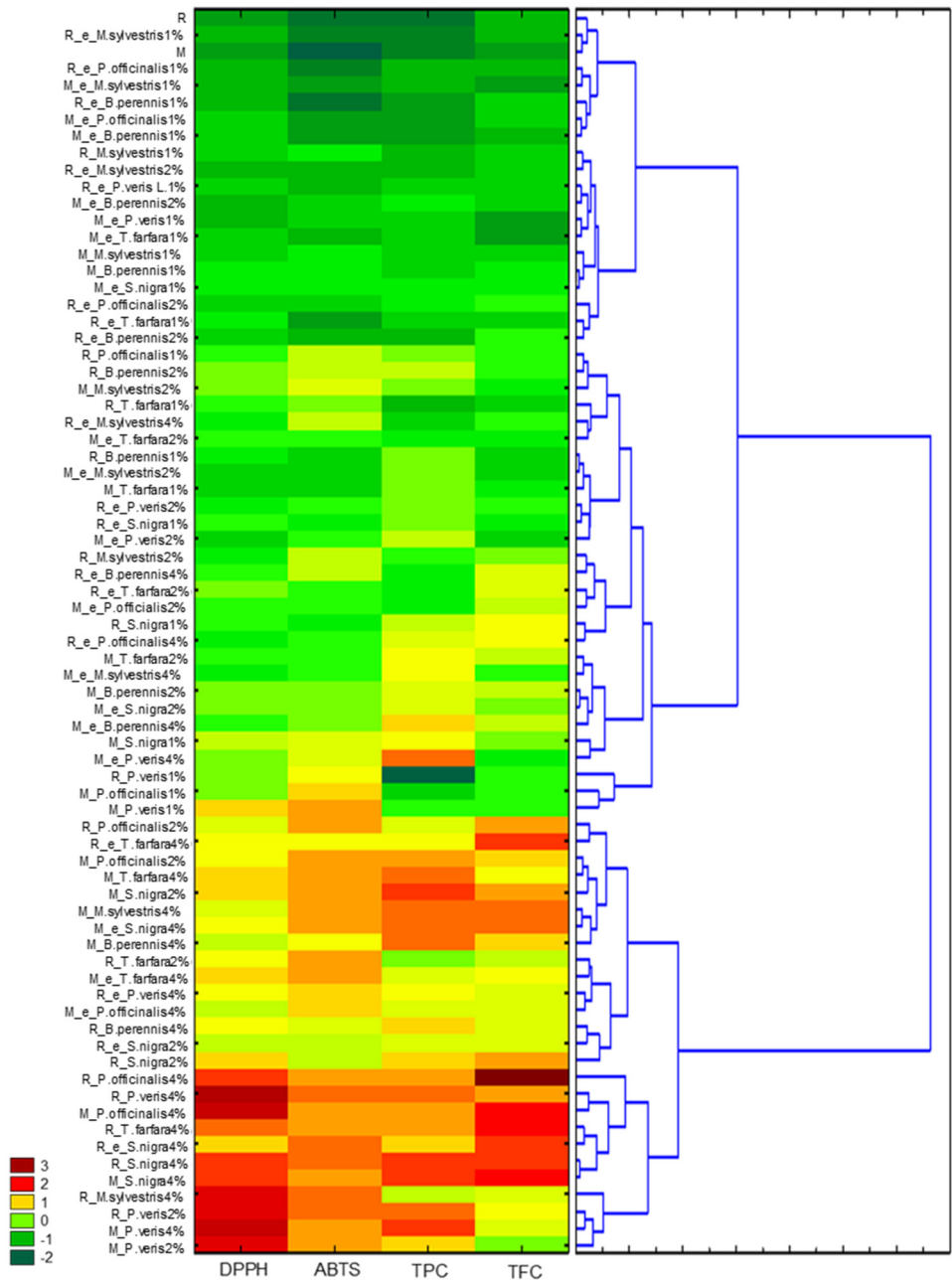


Figure 1. Hierarchical clustering analysis and heatmap visualization of honey enriched by wild-growing spring flowers based on antioxidant activity determined by DPPH and ABTS method, the total content of phenolic compounds (TPC), and the total content of flavonoids (TFC). R—rapeseed honey, M—multifloral honey, R_e—rapessed honey enriched by extract, M_e—multifloral honey enriched by extract, *M. sylvestris*—*Malva sylvestris* L., *P. officinalis*—*Pulmonaria officinalis* L., *B. perensis*—*Bellis perennis* L., *T. farfara*—*Tussilago farfara* L., *P. veris*—*Primula veris* L., *S. nigra*—*Sambucus nigra* L. Cluster analysis was performed using standardized data. The colors on the heat map represent the values of individual parameters, with red indicating high values and dark green indicating low values.

The heat map was used to depict the content of bioactive compounds (polyphenols, flavonoids) in individual samples, as well as their antioxidant activities. This facilitated the identification of samples with the highest health-promoting potential. The analysis was conducted using the Euclidean distance as the measure of distance and the Ward's method as the method of linking objects. The variable importance was determined based on the C&RT model. The obtained predictor importance values are as follows: 1 (DPPH), 0.86 (ABTS), 0.76 (TPC), 0.68 (TFC). The analyzed samples were divided into three main clusters. In cluster analysis, samples with similar values of analyzed variables are placed close to each other. It can be observed that one cluster stands out (encompassing samples with the highest content of phenolic compounds and flavonoids, consequently exhibiting the highest antioxidant activity). Samples in the remaining clusters generally exhibited lower values, with few exceptions, such as M_e_P.veris 4% (multi-floral honey enriched with a 4% addition of extract from *Primula veris* L.), showing higher values in terms of TPC. Analyzing the obtained results, it can be observed that the effectiveness of enrichment in bioactive compounds depends primarily on the plant species. *P. veris* and *S. nigra* stand out, particularly. The form of the additive is also crucial—the dried form proved to be significantly more effective than the extract. Furthermore, as anticipated, the enrichment effectiveness increases with the applied dose (when separately analyzing the dried form and the extract, respectively). Interestingly, the impact of the honey variety used as the matrix for enrichment was not as significant. A notable case is the honey enriched with dried *P. veris*, which exhibited very high activity in the DPPH test, a relatively high content of phenolic compounds, but a low level of flavonoids. This may indicate that, in the case of this plant species, other compounds are responsible for the antioxidant activity against DPPH, which were not detected in the analyzed spectrophotometric tests. Further studies require a more in-depth analysis considering the content of specific phenolic compounds and the identification of other bioactive compounds.

4. Conclusions

This report proved that the application of two plant forms has enriched the content of phenolic compounds within honey and its antioxidant capacity. Furthermore, rapeseed-type honey produces a better matrix for the incorporation of natural plant metabolites into honey. More detailed analysis of modified types of honey is needed to determine phenolic compound profiles and honey effects on some bacterial and yeast pathogens. On the other hand, the high sugar content in honey suggests that a comparison of the glycemic indices of clear and enriched honeys is necessary.

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Data Availability Statement: The data and samples of the compounds presented in this study are available on request from the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

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