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# Sourcing New Ingredients for Organic Cosmetics: Phytochemicals of *Filipendula vulgaris* Flower Extracts

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**Abstract:** Plant extracts are well known for their beneficial cosmetic properties based on bioactive phytochemicals with highly demonstrated antimicrobial and antioxidant capacities as phenolic compounds. This work presents the polyphenolic characterization of different extracts from organically grown *Filipendula vulgaris*. An ultrasound-assisted extraction procedure is proposed for obtaining ethanolic extracts at the laboratory level, both from dried and fresh plants, to be compared with those obtained by classical flower processing techniques for cosmetic purposes. The individual quantification of target phenolic compounds was carried out using liquid chromatography-mass spectrometry (LC-MS/MS). The results revealed the presence of 24 phenolic compounds in the analyzed samples, ranging in concentrations from 0.1 to 71.64 μg $g^{-1}$. The total phenolic content (TPC) ranging from 1163 to 6114 mg GAE $L^{-1}$, and the antioxidant activity (AA), from 6 to 52 mmol TRE $L^{-1}$, were also evaluated. The differences were established between the plant material and extraction technique.

**Keywords:** polyphenols; plant extracts; ultrasound-assisted extraction; total phenolic content; antioxidant activity; liquid chromatography; tandem mass spectrometry



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## 1. Introduction

In recent years, social demand for greener and more sustainable trends in cosmetic formulations using natural origin ingredients has been increasing. Plant extracts are traditionally used as a good source of natural ingredients, and new scientific evidence confirms this fact [1]. In addition, the movement towards circular economy schemes is becoming prevalent. In this sense, extraction alternatives are emerging to recover and reuse byproducts from the agri-food and forestry industries [2–5]. This innovative concept involves the alternative uses of both coproducts and subproducts generated in commercial exploitation, as well as using plants previously cultivated for other purposes (for spices and infusions) or even other wild plants with untapped uses. Thus, the application of plant-derived byproducts as ingredients in cosmetics is a current trend [6]. Indeed, if these residues come from organic agriculture, they constitute an even more valuable resource.

*Filipendula vulgaris* is a perennial plant belonging to the family *Rosaceae* with pinkish-white flowers, growing wild in Europe, Asia, and northwest Africa, mainly on dry non-acidic grasslands and sunny slopes [7]. Different parts of this plant (including flowers, stems, and underground organs such as rhizomes with tuberous roots) have been used as raw materials in traditional medicine [8], given the rich phenolic content of this genus [9] and the evidence of anti-inflammatory, anti-rheumatic, and diuretic activity. In some cases, a certain beneficial property is associated with a specific plant part [10]. More recently, new aspects of their biological activity, such as their antioxidant capacity, have become

the subject of scientific interest [11]. *Filipendula vulgaris* is an important source of different secondary metabolites that can act as chemopreventive agents with beneficial activities for human health [12]. Due to the dynamic changes in agricultural land use, the native habitats of this species are gradually disappearing, so the decrease in natural resources requires its introduction into cultivation [13] and, clearly, for environmental concerns, in line with the necessary reduction in the use of synthetic fertilizers and phytosanitary products and the introduction of sustainable crops, organic farming is the best option.

Different parts of the Filipendula plant including the leaves and underground organs but not the flowers, have been considered for polyphenolic analysis using high performance liquid chromatography with a UV-Vis detector (HPLC-UV) [14]. Lyophilized flower infusions have been evaluated for their antioxidant and anti-inflammatory properties and their active components by HPLC with a diode array detector (DAD) [15]. HPLC-DAD was also used to determine phenolic compounds in methanolic extracts of above-ground and underground organs from both fresh and dried cultivated Filipendula plants [16]. Finally, in addition to exploring the anti-inflammatory effects and antioxidant properties, two research papers introduced mass spectrometry in the analysis of methanolic extracts of air-dried aerial parts and roots obtained by maceration [17] and extracts isolated by different extraction techniques and solvents depending on the plant material [18].

Plant extracts can be obtained using classical methods such as hydrodistillation, but other techniques including extraction and maceration are also frequently used. The International Organization for Standardization (ISO) adopts definitions for alcoholate and hydrolate [19]. While alcoholate is defined as a distillate resulting from the distillation of natural raw material (plant origin) in the presence of ethanol in varying concentrations, hydrolate is an aqueous distillate remaining after the steam distillation and separation of the essential oil whenever possible.

The goal of this work is the investigation of the polyphenolic composition of different extracts of *Filipendula vulgaris* flowers, both obtained on an industrial level (hydrolates and alcoholates) and on a laboratory scale (See Figure 1). For this study, plants organically grown in Galicia (NW Spain) were considered as the raw materials from which to obtain extracts to be used as ingredients in cosmetics, providing them with an organic seal. A green, fast, and low-cost sample preparation strategy based on ultrasound-assisted extraction (UAE) was assessed for obtaining extracts from fresh, dried, and frozen plants at the laboratory level. The quantification of the phenolic compounds was carried out using liquid chromatography-tandem mass spectrometry (LC-MS/MS). The total polyphenolic content (TPC) index and the antioxidant activity (AA) of the extracts were also evaluated. Finally, the main compositional differences found between the extracts obtained by the laboratory-optimized procedure and the hydrolates and alcoholates are discussed.

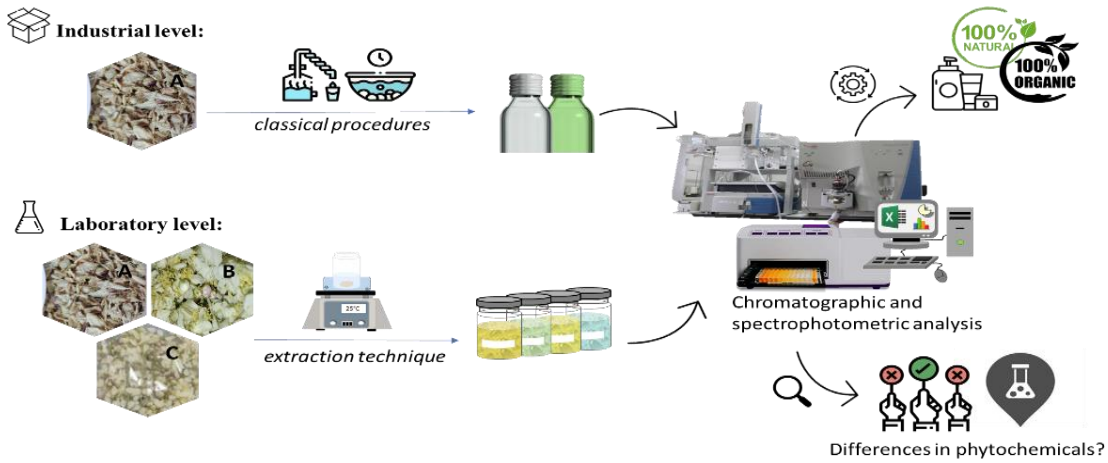


**Figure 1.** Proposed work approach: dried (**A**), fresh (**B**), and frozen (**C**) samples.

Thus, the novelty of this work lies in the characterization of different extracts of *Filipendula vulgaris* to obtain detailed information on their phenolic chemical profile as well as their bioactivities for cosmetic purposes.

## 2. Materials and Methods

### *2.1. Chemicals, Reagents, and Materials*

The target phenolic compounds, their CAS numbers, molecular masses, retention times, and MS/MS transitions are summarized in Supplementary Table S1. MS-grade ethanol and ultrapure water were supplied by Scharlab (Barcelona, Spain). MS-grade methanol was obtained from Sigma Aldrich Chemie GmbH (Steinheim, Germany) and formic acid was obtained from Merck (Darmstadt, Germany). Folin–Ciocalteu's phenol reagent and 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox®) were purchased from Sigma Aldrich, 2,2-diphenyil-1-picrylhydrazyl (DPPH) was supplied by TCI (Tokyo Chemical Industry) (Tokyo, Japan) and sodium carbonate ($Na_2CO_3$) was supplied by Panreac (Barcelona, Spain). All chemicals and reagents were of analytical grade.

### *2.2. Plant Material*

Organically grown Filipendula vulgaris flowers fresh samples, the corresponding dried samples after drying in an industrial oven at a temperature of 35 °C, and the derived hydrolates and alcoholates were supplied by Milhulloa (S. Coop. Galega, Galicia, Spain). Filipendula flowers were collected from the Palas de Rei locality (UTM 29T585314 47484481). Taxonomic identification was performed at the Department of Forest Ecosystems, Lourizán Forest Research Centre (Pontevedra, Galicia), and voucher specimens (71925 LOU-071 925 and 71,925 LOU-071 9256) were deposited in the LOU Herbarium of that centre. Hydrolates were obtained using the classical steam distillation procedure, and alcoholates were produced by maceration in organic wheat alcohol for seven days and then filtered. In both cases, the raw material was dried flowers.

Fresh flowers were processed immediately after collection, and the remaining samples were frozen, with the aim of repeating the extraction of the frozen sample after one week. Dried samples were kept in the original paper bags in a non-humid place until their analysis. Obtained extracts were stored and protected from light.

### *2.3. Sample Preparation*

Before the extraction process, Filipendula samples (fresh, dried, or frozen) were ground in a glass mortar to facilitate their breakup. Afterwards, a 0.200 g sample was exactly weighed into a 10 mL glass vial and 2 mL of ethanol was added. The vial was sealed with an aluminum cap furnished with PTFE (polytetrafluoroethylene)-faced septa and immersed in an ultrasound bath (US, J Selecta, Barcelona, Spain) for 10 min at 50 kHz and room temperature. The extract was then filtered through 0.22 µm PTFE filters. Two extraction replicates as well as an extraction blank were performed for each sample. All solutions were stored at −20 °C until analysis. The experimental procedure is summarized in Figure 2.

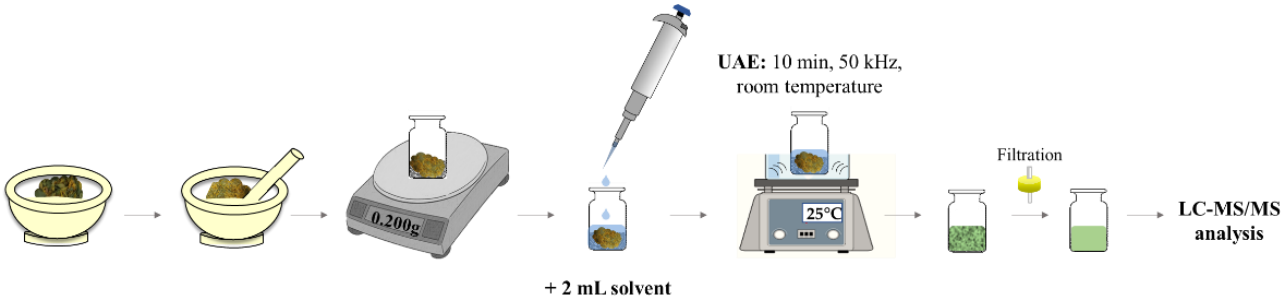


**Figure 2.** Schematic representation of the UAE experimental procedure.

### 2.4. Determination of TPC

The total polyphenolic content (TPC) of the extracts was determined according to the Folin–Ciocalteu (FC) colorimetric method described by Singleton and Rosssi [20] and following Zhang's guidelines for microtitration in 96-well plates [21]. Briefly, a total of 20 µL of the diluted extract was mixed with 100 µL of FC reagent (1:10, $v/v$) and 80 µL of sodium carbonate solution (7.5 g $L^{-1}$). The mixture was shaken and isolated in the dark for 30 min and then the absorbance at 760 nm was measured in a microplate reader (BMG LABTECH, Ortenberg, Germany). The TPC was quantified by a calibration curve prepared with gallic acid standard solutions covering a concentration range of 20 to 160 mg $L^{-1}$ (0.200–0.800 abs, $R^2$ = 0.9986). TPC was expressed as mg of gallic acid equivalent (GAE) per liter of extract (mg GAE $L^{-1}$).

### 2.5. Determination of AA

The antioxidant activity (AA) of the extracts was determined using the DPPH method [22], as described by Symes et al. [23]. Briefly, 100 µL of the extract at eight different concentration levels was placed in a 96-well plate and mixed with 100 µL of DPPH reagent (0.25 mM) prepared in methanol. The mixture was kept in the dark for 10 min and the absorbance measurement was performed at 515 nm. The AA was quantified using a calibration curve prepared with Trolox® (3 to 31 mg $L^{-1}$ (0.200 to 0.800 abs), $R^2$ = 0.9984). The AA was expressed as millimoles of Trolox® equivalent (TRE) per liter of extract (mmol TRE $L^{-1}$).

### 2.6. LC-MS/MS Analysis

Quantification of the individual polyphenols was performed by LC-MS/MS employing a Thermo Scientific (San José, CA, USA) instrument based on a TSQ Quantum Ultra™ triple quadrupole mass spectrometer equipped with a heated electrospray ionization (HESI) source, and an Accela Open autosampler with a 20 µL loop. The optimal instrumental conditions for the detection of the target phenols were adapted from those previously optimized by Celeiro et al. [2]. The chromatographic separation was achieved on a Kinetex C18 column (2.6 µm, 100 × 2.1 mm) with a guard column (SecurityGuardTM ULTRA Holder) obtained from Phenomenex (Torrance, CA, USA). The injection volume was 10 µL and the column temperature was set at 50 °C. The mobile phase consisted of water (A) and methanol (B), both containing 0.1% formic acid. The eluted program started with 5% of B (held for 5 min), going up to 90% of B over 11 min (held for 3 min). Then, a return to initial conditions was achieved in 6 min. The mobile phase flow rate was 200 µL $min^{-1}$. The total run time for each injection was 20 min. The mass spectrometer and the HESI-II source were working simultaneously in the positive and negative modes (see ionization mode for each target compound in Table S1). Selected reaction monitoring (SRM) acquisition mode was implemented monitoring 2 or 3 transitions per compound (see Table S1), for an unequivocal identification and quantification of the target compounds. Additionally, new compounds were added to the optimized method [2], reaching a total of 55 analyzed compounds. The MS/MS parameters for the new compounds were optimized by individual direct infusion, and the most abundant collision-induced fragments were considered for quantification. The system was operated using Xcalibur 2.2 and Trace Finder™ 3.2. In all cases, good linearity with coefficients of determination ($R^2$) was obtained (see Table S1).

## 3. Results and Discussion

### 3.1. TPC and AA

TPC and AA for the extracts considered (hydrolate, alcoholate, and ultrasound-obtained extracts of fresh, frozen, and dry flowers) were measured. The obtained values are summarized in Table 1.

**Table 1.** Mean TPC and AA values for the studied extracts: alcoholate, ultrasonic extracts from fresh, frozen, and dried samples.

| Extract | DF [a] | TPC (mg GAE $L^{-1}$) | DF | AA (mmol TRE $L^{-1}$) |
|---|---|---|---|---|
| Alcoholate | 16 | 1163 ± 50 | 128 | 6 ± 1 |
| Fresh sample extract | 60 | 3599 ± 82 | 1024 | 52 ± 3 |
| Frozen sample extract | 60 | 3034 ± 21 | 512 | 35 ± 2 |
| Dried sample extract | 64 | 6114 ± 24 | 1024 | 45 ± 4 |

[a] DF: Dilution factor.

The TPC and AA values of the pure hydrolate (without dilution) were not quantifiable. The TPC values obtained for the fresh and frozen sample ultrasonic extracts were similar, around 3000 mg GAE $L^{-1}$. Notably, about twice that value was obtained for the dried sample, showing that the dried flower extract had the highest TPC value. However, the results were different for the AA. The fresh sample showed an AA close to that of the dried sample, both higher than the frozen sample. A high correlation between the TPC and AA indexes have been reported in certain cases [24,25], but this correlation is not so evident as in other studies [26,27] due to the different antioxidant activity of the polyphenols, as well as their breakdown and reactivity. Alcoholate exhibited lower values of TPC and AA than the ultrasonic extracts.

### *3.2. Chromatographic Analysis*

LC-MS/MS analysis revealed that the studied F. vulgaris extracts contain several classes of phenolic compounds, mainly different types of flavonoids, but also phenolic acids and aldehydes. A total of 24 individual phenolic compounds were identified, and their quantification is detailed in Table 2. As can be seen, the presence in the extracts of quercetin, astragalin, gallic acid and kaempferol stands out.

**Table 2.** Mean concentration ($n$ = 3) of phenolic compounds (μg $mL^{-1}$) found in the extracts of Filipendula vulgaris (concentrations > 10 ppm are marked in bold).

| | Mean Concentration (μg $mL^{-1}$) | | | |
|---|---|---|---|---|
| **Phenolic Compound** | **Alcoholate** | **Fresh Sample Extract** | **Frozen Sample Extract** | **Dried Sample Extract** |
| Gallic acid | **20.97 ± 1.22** | 7.60 ± 0.03 | 6.03 ± 0.34 | **63.17 ± 4.02** |
| Phloroglucinic acid | 1.29 ± 0.51 | 0.53 ± 0.12 | 0.41 ± 0.02 | 4.22 ± 0.17 |
| 3,4-dihydroxybenzaldehyde | N.D. | N.D. | N.D. | 1.40 ± 0.004 |
| Gentisic acid | N.D. | N.D. | N.D. | 0.24 ± 0.03 |
| 3-hydroxybenzoic acid 4-hydroxybenzoic acid [a] | 0.71 ± 0.06 | N.D. | N.D. | N.D. |
| Procyanidines B1, B2 and C1 [a] | 0.09 ± 0.005 | 0.85 ± 0.02 | 0.33 ± 0.04 | 0.88 ± 0.08 |
| Catechin | 1.40 ± 0.08 | 7.51 ± 0.01 | 3.60 ± 0.03 | 2.91 ± 0.01 |
| Resorcylic acids [b] | 0.57 ± 0.01 | 0.91 ± 0.05 | 0.73 ± 0.02 | 1.09 ± 0.02 |
| 4-Hydroxybenzaldehyde | 0.29 ± 0.001 | N.D. | N.D. | N.D. |
| Caffeic acid | 0.49 ± 0.002 | 0.43 ± 0.02 | 0.43 ± 0.01 | 0.50 ± 0.001 |
| 4-hydroxycinnamic acid | 0.69 ± 0.001 | N.D. | N.D. | 0.88 ± 0.01 |
| Quercetin-3-glucuronide | N.D. | N.D. | N.D. | 1.18 ± 0.03 |
| Quercetin-3-glucoside | N.D. | N.D. | N.D. | **71.64 ± 0.17** |
| Ellagic acid | 2.74 ± 0.04 | 6.53 ± 0.22 | 4.48 ± 0.19 | 8.62 ± 0.04 |
| Rosmarinic acid | 0.14 ± 0.002 | N.D. | N.D. | 0.17 ± 0.004 |

**Table 2.** *Cont.*

| | Mean Concentration (μg mL$^{-1}$) | | | |
|---|---|---|---|---|
| **Phenolic Compound** | **Alcoholate** | **Fresh Sample Extract** | **Frozen Sample Extract** | **Dried Sample Extract** |
| Astragalin | **11.26 ± 0.25** | **40.80 ± 0.43** | **39.32 ± 0.45** | **17.42 ± 0.39** |
| Quercetin | **37.72 ± 2.98** | **33.24 ± 0.27** | **38.14 ± 0.07** | **46.14 ± 0.90** |
| Naringenin | 0.45 ± 0.02 | 0.34 ± 0.005 | 1.19 ± 0.02 | 0.63 ± 0.001 |
| Luteolin | 0.95 ± 0.06 | 1.09 ± 0.05 | 1.57 ± 0.06 | 1.13 ± 0.03 |
| Kaempferol | **10.57 ± 0.79** | 9.46 ± 0.14 | **15.63 ± 0.23** | **12.31 ± 0.05** |

[a] Quantified as the sum of the individual compounds. [b] Quantified as the sum of the individual isomers (γ and α). N.D.: not detected.

The highest number of polyphenols was detected in the dried flower extract, which, in general, also showed the highest concentrations of the compounds, as can be seen in Figure 3. This stacked column chart has been constructed with the concentration values in μg mL$^{-1}$ and shows the relative contribution (%) of the polyphenol concentration of each sample subgroup (indicated by colors) for individual compounds.

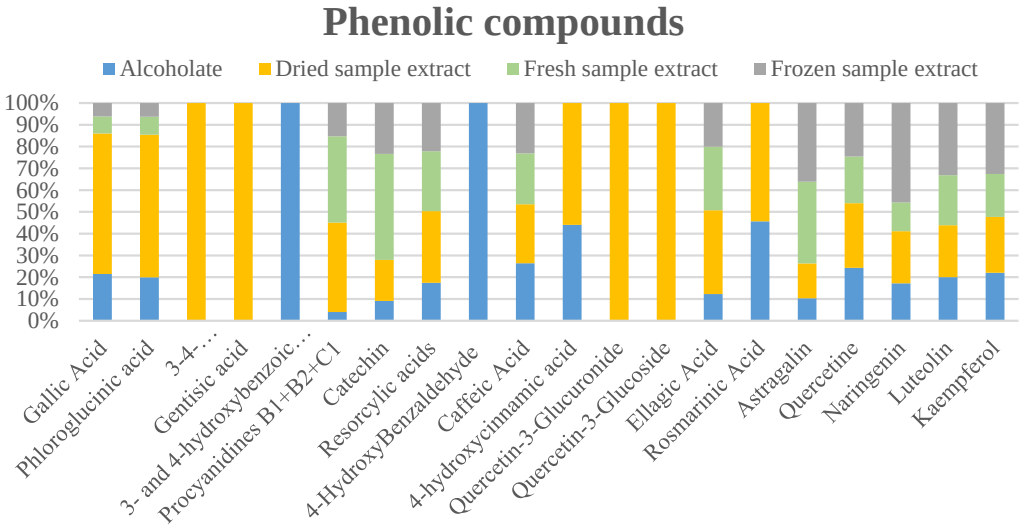


**Figure 3.** Distribution of polyphenols in the different samples studied. Resorcylic acids include two isomers: α and γ.

Figure 4 summarises the results for the determination of polyphenols in the different extracts. The most significant compounds in the alcoholate were quercetin, gallic acid, astragalin, and kaempferol, all at concentrations above 10 ppm (see Table 2). The 3- and 4-hydroxybenzoic acids and 4-hydroxybenzaldehyde were only detected in this sample. The composition of the extracts obtained from the fresh and frozen flower samples is quite similar. Of note is the high concentration of quercetin-3-glucoside in the dry sample extract, a compound that was identified exclusively in this extract.

As indicated above, the polyphenolic content of the hydrolate was negligible. Their use in cosmetics is not related to their polyphenol content, but to their content of volatile compounds and thus to their flavouring properties.

The results of the LC-MS/MS determination consistently support the values obtained for the spectrophotometric indices (TPC and AA). In general, the concentration of polyphenols in the alcoholate was the lowest, which explains its lower TPC and AA values. Similarly, the higher number of compounds identified and their higher concentrations in the dry extract of Filipendula are consistent with the higher TPC and AA values of this extract. The contribution of some specific polyphenols in higher concentrations in the fresh flower extract, such as catechin or astragalin, could explain its higher antioxidant activity. In this

regard, it is important to note that structure–activity relationships play a very important role in determining whether compounds will exhibit an antioxidant effect [28,29]. Therefore, the AA index values can vary significantly between different classes of compounds, even between compounds of the same type [30].

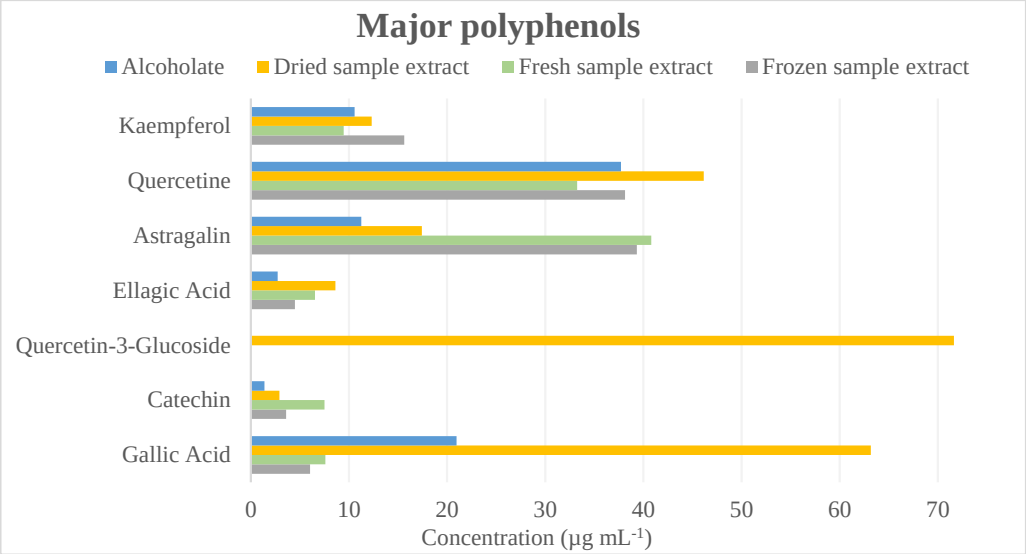


**Figure 4.** Major polyphenols found in the Filipendula extracts.

Five of the compounds identified in Filipendula extracts in this work, namely gallic and ellagic phenolic acids, catechin, quercetin, and astragalin, have also been found in the methanolic extracts of Filipendula underground organ air-dried samples at different stages of development [14]. The presence of gallic acid, ellagic acid, and astragalin, as well as quercetin-3-glucoside, was also determined in the infusions of the freeze-dried flowers of Filipendula vulgaris [15], coinciding with the results of the present study in the higher concentration of quercetin-3-glucoside. It is significant that each compound isolated from a particular part of the Filipendula plant has specific biological activities associated with it [10]. Another study involving the flowers, in addition to the phenolics mentioned above, confirmed the presence of kaempferol [16]. Mass spectrometry was used in two papers, the first of which positively identified 11 phenolic compounds, and tentatively identified other related substances [17], confirming seven of the compounds identified in the present work. The second paper concluded that the quantitative distribution of the compounds identified in the extracts depended on the extraction solvent applied, but in general, the highest efficiency corresponded to the methanolic extract [18], in which a total of 18 polyphenolic compounds and derivatives were determined, four of them coinciding with those determined in this work (gallic acid, catechin, ellagic acid, and astragalin). However, to the authors' knowledge, the presence of other polyphenols, such as hydroxybenzoic acids or procyanidins, has never been reported in flower extracts. 3-hydroxybenzoic acid is used in cosmetics as a skin conditioning agent, while 4-hydroxybenzoic acid has properties as a fragrance and preservative agent (data obtained from PubChem). As for the procyanidin oligomers determined in the present work, they have been investigated as potential hair growth-promoting agents [31].

In summary, F. vulgaris contains a wide range of polyphenols with health benefits and, consequently, extracts derived from the plant can be considered a promising source of ingredients for use in cosmetics based on natural ingredients and marketed under the organic cosmetics label.

## 4. Conclusions

The research carried out in this study provided information on the phytochemical characterization of *Filipendula vulgaris* flowers grown organically in the region of A Ul-

loa (Galicia). The extracts were shown to contain a high number of polyphenols, and in significant concentrations in some cases. The slight differences found in the bioactivities and polyphenolic composition between the different types of extracts were associated with the processing procedures. The low-temperature drying process generally used to process and store botanical species is suitable for preserving and concentrating their bioactive polyphenol content. Furthermore, the results obtained suggest that the inclusion of an ultrasound-assisted extraction step could increase the extractive efficiency of some substances of interest and that this technique is compatible with organic cosmetics criteria. LC-MS/MS analysis allowed the identification and quantification of 24 phenolic compounds, highlighting the concentration of some compounds at levels of up to 30 μg $mL^{-1}$. Most of the identified compounds have demonstrated interesting bioactive properties, such as antioxidant and antimicrobial properties, which makes these extracts a consideration for use in the first cosmetic prototypes currently under development.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/cosmetics9060132/s1, Table S1: Phenolic compounds identified: CAS number, molecular mass (Mm), retention times (Rt), ionization mode, MS/MS transitions, and coefficients of determination ($R^2$).

**Author Contributions:** Conceptualization, L.R., M.L. and C.G.-J.; methodology, L.R.; software, L.R.; validation, L.R., M.L. and C.G.-J.; formal analysis, L.R., M.L. and C.G.-J.; investigation, L.R.; resources, M.L. and C.G.-J.; data curation, L.R., M.L. and C.G.-J.; writing—original draft preparation, L.R.; writing—review and editing, M.L. and C.G.-J.; visualization, M.d.C.V., M.J.E., M.L. and C.G.-J.; supervision, M.L. and C.G.-J.; project administration, M.d.C.V., M.J.E., M.L. and C.G.-J.; funding acquisition, M.d.C.V., M.J.E., M.L. and C.G.-J. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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