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# Utilizing Herbal Dust: Ultrasound-Assisted Extraction of Green Tea, Hibiscus, and Lemon Balm Filter Tea Industry Waste

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**Abstract:** The rise of the global tea industry market, influenced by the growing demands for healthier diet options, resulted in the constant increase in herbal tea production. In accordance, increased production leads to increased waste generation, especially in the area of filter tea production, which generates waste in the form of powdered plant material with particle sizes lower than 0.315 mm. The generated amount of this powdered plant material, also called herbal dust, can vary in the range from 10 to 40% of the total processed plant, and it is often considered waste only due to its size. Therefore, within this study, ultrasound-assisted extraction (UAE) was utilized for the extraction of green tea (*Camellia sinensis* L.), lemon balm (*Melissa officinalis* L.), and hibiscus (*Hibiscus sabdariffa* L.) herbal dust, and the quality of the obtained extracts was evaluated in terms of total phenolic content (TPC) and phenolic profile. In addition, UAE was conducted on the three different amplitudes (20, 60, and 100%) and two different extraction times (5 and 10 min) in order to investigate and compare the influence of different extraction parameters. The results showed that the maximum TPC for green tea, hibiscus, and lemon balm herbal dust was 152.91 ± 0.74, 60.63 ± 0.10, and 356.22 ± 3.13 mg GAE/g DE, respectively. HPLC analysis conducted for all of the obtained extracts confirmed the presence of several phenolic compounds, with the highest concentrations of epigallocatechin gallate (EGCG) for the extracts of green tea, and neochlorogenic acid for hibiscus herbal dust extracts. The HPLC analysis of the lemon balm extracts recorded the highest concentration of rosmarinic acid for all of the UAE conditions. The results reported within this study indicate that previously considered waste, herbal dust plant material can be successfully extracted by application of UAE and that the obtained extracts exhibit concentrations of bioactive compounds comparable to the extracts of the commercially available plant material.

**Keywords:** waste valorization; ultrasound-assisted extraction; green tea; hibiscus; lemon balm; polyphenols



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## 1. Introduction

With the increasing awareness about the importance of a healthier, low-sugar, natural-based diet, the world's demand for herbal teas is on the constant rise. According to the available data, in 2022, the global value of the tea industry was around 112.2 billion dollars, which amounted to about 6.7 million tons of consumed dried plant material from both *Camellia sinensis* L. plant as well as other plants with proven biological activity. It is expected that the tea industry will reach an annual production of 7.4 million tons by the year 2025 [1]. In accordance, increased production of herbal teas results in generating larger amounts of waste which occurs as a residue after processing plant material, especially in the area of filter tea production. During filter tea production, plant material requires grounding

procedures that provide particles in the range from 2.00 to 0.315 mm, known as the "fine cuts". However, this procedure also generates substantial quantities of material with particle sizes less than 0.315 mm in the form of fine powder, which is generally considered waste. This fine powder, or so-called "herbal dust", represents a fraction that cannot be further used for the production of filter tea bags only due to their size and is often discarded [2,3]. The amount of this herbal dust varies depending on the type and species of plant material that is used, and it can vary in the range from 10 to 40% of the total processed material [3]. Given the availability, quality, and quantity of the herbal dust that is generated, the potential for additional utilization of this material is evident. For this study, the quality of the extracts prepared from the herbal dust obtained from the production of green tea (*Camellia sinensis* L.), lemon balm (*Melissa officinalis* L.), and hibiscus (*Hibiscus sabdariffa* L.) was evaluated.

Green tea is one of the most consumed non-alcoholic beverages worldwide, and is produced from dried *Camellia sinensis* L. leaves, originating from the Far East, mainly China and India [4]. Beginning in the 19th century, extensive research proved that green tea, or the non-fermented and dried form of *C. sinensis*, contains more than 400 different organic compounds, many of them being biologically active. Among those compounds, green tea polyphenols are the most abundant and accredited with the most favorable bioactivity. Green tea polyphenols can make up to 35% of green tea leaves and are mostly composed of catechins, but also flavonoids, anthocyanins, and phenolic acids [4]. In addition, green tea also contains flavonoids as well as alkaloids, specifically caffeine (from 2 to 5%), theophylline, and theobromine [5]. Accordingly, recent studies reported that green tea extracts (mostly methanolic or ethanolic) exhibit antioxidant, antimicrobial, antibacterial, antiviral, and antitumor activity [6].

*Melissa officinalis* L., commonly known as lemon balm, is one of the plants that have been used in traditional medicine since ancient times. This perennial, lemon-scented plant belonging to the mint family (*Lamiaceae*) is a native species in Europe, Central Asia, and Iran, and mainly inhabits sandy and low shrub-covered areas [7]. Traditionally, lemon balm was used for calming and anti-melancholy, improvement of memory, mood, and sleep, as well as treatment of dysentery, intestinal ulcers, toothache, and morning sickness. This wide application of lemon balm can be attributed to its diverse phytochemical composition which comprises mainly volatile compounds (geranial, neral, and geraniol), phenolic compounds (rosmarinic acid, gallic acid, and salvianolic acid), and flavonoids (luteolin, apigenin, rutin, catechin, and quercetin). Although volatile compounds or essential oils are considered to be the main carriers of biological activity, polyphenols also contribute to the overall therapeutic effects of lemon balm [8]. Therefore, modern medicine reports several health-beneficial activities connected to the presence of phenolic compounds, such as neuroprotective, cardiovascular, cytotoxic, anti-inflammatory, hypoglycemic, antioxidant, antimicrobial, and antispasmodic effects [9].

The genus Hibiscus includes more than 300 species of perennial plants, shrubs, or trees, distributed in tropical and subtropical regions of the world [10]. Within this genus, one distinctive species originating from Africa, *Hibiscus sabdariffa* L., also known as roselle, has a worldwide application in the production of herbal teas, as well as a long history of traditional therapeutic usage [10]. Traditionally, *H. sabdariffa* has been used for its diuretic and cardiovascular effects, and in many African countries, it has been used for wound treatment, lowering body temperature, as well as a remedy for sore throats and coughs [11]. These therapeutic properties are mainly attributed to the presence of organic acids such as ascorbic acid, maleic acid, or oxalic acid, as well as phenolic compounds, mainly flavonoids (quercetin or myricetin) and anthocyanins (delphinidine-3-sambubioside and cyanidine-3-sambubioside). In accordance, many studies report that *H. sabdariffa* exhibits antihypertensive, anti-inflammatory, antibacterial, neuroprotective, and hepatoprotective activity [10,11].

Following the principles of green chemistry within this study, ultrasound-assisted extraction (UAE) was used to determine the quality of the herbal dust extracts of green tea

(*Camellia sinensis* L.), lemon balm (*Melissa officinalis* L.), and hibiscus (*Hibiscus sabdariffa* L.). UAE has been widely used for the extraction of different plant materials, and it was chosen for this study as a proven, green, and cost- and time-effective technique for the efficient extraction of plant-based phenolic compounds. The quality of the extracts obtained by UAE was evaluated in terms of the total phenolic content (TPC) and phenolic profile, which was determined by HPLC analysis. Additionally, to utilize the herbal dust to its absolute full potential, cellulosic material that was left over after the extraction was collected and incinerated to create biochar or herbal dust ash, which was integrated into rubber as a filler compound. The detailed biochar characteristics, as well as the procedure of biochar creation, analysis, and integration into the final product from the material that was left over after the UAE will be covered in the subsequent study, while this study reports on the results of the extraction itself.

## 2. Materials and Methods

### 2.1. Plant Material and Chemicals

Herbal dusts of *Camellia sinensis* leaves, *Melissa officinalis* leaves, and *Hibiscus sabdariffa* calyx were obtained from the company Fructus (Bačka Palanka, Serbia), a local tea manufacturer. All of the plant samples were material that was left after the production of herbal tea preparations. The moisture content of plant material was 6.67, 8.07, and 7.83% for green tea herbal dust, hibiscus herbal dust, and lemon balm herbal dust, respectively. Particle size for all of the plant materials was <0.35 mm and therefore considered herbal dust. Folin–Ciocalteu reagent was obtained from Institute Mol (Stara Pazova, Serbia). The solvent used for the UAE extraction was diluted 96% ethanol, obtained from Centohem (Šabac, Serbia). For the HPLC analysis, ultrapure water was obtained by the Millipore Simplicity 185 system (Darmstadt, Germany), while the methanol and formic acid were of HPLC grade and were obtained from J.T. Baker (Phillipsburg, NY, USA). All HPLC standards were calculated to 100% purity.

### 2.2. Ultrasound-Assisted Extraction

Ultrasound-assisted extraction (UAE) was performed by ultrasonic probe UP200Ht with a working frequency of 26 kHz and a maximum power output of 200 W (Sonotrode S26d7, produced by Hielscher GmbH, Teltow, Germany). For all of the plant materials (*Camelia sinensis*, *Melissa officinalis*, and *Hibiscus sabdariffa*), the extractions were performed at three different amplitudes (20, 60, and 100%) and two different times of extraction (5 and 10 min). The 50% aqueous ethanol solution was used for all of the extractions, and the ratio of plant material to solvent (S/L ratio) was 1:20 ($w/v$). The extraction temperature was constantly kept under the 40 °C, using an ice bath (Table 1). For all of the extractions, a magnetic stirrer was used to constantly mix the samples (200 rpm). After the extraction procedure, the cellulosic plant materials were separated from the extracts by vacuum pump filtration (filter paper grade 34 N, 0.20 mm thickness) and stored in plastic bags, at −18 °C, for further analysis. The extracts were additionally filtered through a 0.45 µm nylon syringe filter. After the filtration, the extracts were stored in plastic bottles and kept until further analysis at 4 °C.

**Table 1.** Conditions of UAE for green tea (*C. sinensis*), hibiscus (*H. sabdariffa*), and lemon balm (*M. officinalis*).

| Extract (Green Tea) | Extraction Conditions | Extract (Hibiscus) | Extraction Conditions | Extract (Lemon Balm) | Extraction Conditions |
|---|---|---|---|---|---|
| GT 1 | AMP 100%, 5 min, 50% EtOH, 1:20 | HB 1 | AMP 100%, 5 min, 50% EtOH, 1:20 | LB 1 | AMP 100%, 5 min, 50% EtOH, 1:20 |
| GT 2 | AMP 100%, 10 min, 50% EtOH, 1:20 | HB 2 | AMP 100%, 10 min, 50% EtOH, 1:20 | LB 2 | AMP 100%, 10 min, 50% EtOH, 1:20 |

**Table 1.** *Cont.*

| Extract (Green Tea) | Extraction Conditions | Extract (Hibiscus) | Extraction Conditions | Extract (Lemon Balm) | Extraction Conditions |
|---|---|---|---|---|---|
| GT 3 | AMP 60%, 5 min, 50% EtOH, 1:20 | HB 3 | AMP 60%, 5 min, 50% EtOH, 1:20 | LB 3 | AMP 60%, 5 min, 50% EtOH, 1:20 |
| GT 4 | AMP 60%, 10 min, 50% EtOH, 1:20 | HB 4 | AMP 60%, 10 min, 50% EtOH, 1:20 | LB 4 | AMP 60%, 10 min, 50% EtOH, 1:20 |
| GT 5 | AMP 20%, 5 min, 50% EtOH, 1:20 | HB 5 | AMP 20%, 5 min, 50% EtOH, 1:20 | LB 5 | AMP 20%, 5 min, 50% EtOH, 1:20 |
| GT 6 | AMP 20%, 10 min, 50% EtOH, 1:20 | HB 6 | AMP 20%, 10 min, 50% EtOH, 1:20 | LB 6 | AMP 20%, 10 min, 50% EtOH, 1:20 |

### *2.3. Determination of the Extraction Yield*

Yield of the extraction represents the total mass of all extracted components. For all of the obtained UAE extracts, the yield was determined by evaporating solvent using a stage evaporation procedure. Firstly, the solvent was evaporated from the sample by vacuum rotary evaporator and, after that, samples were dried for 3 h at 105 °C, until the constant mass. The mass of dried extract (DE) was evaluated by the standard gravimetric procedure (Table 2). For the yield determination, 5 mL of extract was used, and the results were expressed as mg of dried extract per mL of extract (mg DE/mL).

**Table 2.** Yield of UAE [mg DE/mL].

| Extract | Yield | Extract | Yield | Extract | Yield |
|---|---|---|---|---|---|
| GT 1 | 14.08 | HB 1 | 15.46 | LB 1 | 6.02 |
| GT 2 | 14.16 | HB 2 | 16.28 | LB 2 | 7.42 |
| GT 3 | 14.08 | HB 3 | 16.78 | LB 3 | 5.18 |
| GT 4 | 15.16 | HB 4 | 17.34 | LB 4 | 4.52 |
| GT 5 | 6.32 | HB 5 | 17.16 | LB 5 | 3.4 |
| GT 6 | 9.16 | HB 6 | 16.08 | LB 6 | 4.26 |

### *2.4. Determination of Total Phenolic Content*

The determination of total phenolic compounds in all of the obtained UAE extracts was conducted by the Folin–Ciocalteu method [12]. The absorbance of the test samples was measured at a 750 nm wavelength range, using a single-beam UV/VIS spectrophotometer 6300 Spectrophotometer, Jenway (Staffordshire, UK). The results are expressed as gallic acid equivalents per g of dried extract (mg GAE/g DE). All of the analytic tests were performed in triplicates, and the standard deviation was calculated.

### *2.5. HPLC Analysis of the Polyphenolic Components*

Determination of bioactive components for *Camelia sinensis*, *Melissa officinalis*, and *Hibiscus sabdariffa* UAE extracts was performed by the HPLC (high-performance liquid chromatography) method with UV detection on a Cosmosil 5C18-MS-II column (Nacalai Tesque, Inc., Kyoto, Japan) that was 250 mm long with an internal diameter of 4.6 mm and filled with 5 µm particles. The HPLC system used for the analysis (Agilent, 1260 Infinity II series, Waldbronn, Germany) consisted of a quaternary pump (G7111A), DAD (photo-diode array) detector (G7115A), a column heater with temperature ranges from 10 to 85 °C (G7116A), an autosampler with capacity of 36, 6 mL vials (G7157A), and a fraction collector (G1364E). The system was operated using the computer program ChemStation (Open lab CDS).

The separation of the analyzed compounds was carried out by gradient elution at a flow rate of 1 mL/min for 65 min. In phase A, 0.1% formic acid in ultrapure water (Millipore Simplicity 185 system, Darmstadt, Germany) was used, and in phase B, 0.1% formic acid in methanol was used. The gradient was set as follows: 0.00–8.00 min 90% A; 8.00–16.00 min 75% A; 16.00–28.00 min 55% A: 28.00–55.00 min 20% A; and 55.00–65.00 min 10% A, followed by a period of 15 min where the analysis conditions returned to the initial value. The injection volume was 10 µL, the UV detection wavelength was 240, 250, 260, 270, 280, 330, and 360 nm, and the temperature of the column compartment was 50 °C. The purity of the peaks was calculated using ChemStation software (Open lab CDS), and all analytes were verified through purity factor, spectra threshold, and noise threshold.

### 2.6. Statistical Analysis

All analyses were conducted in triplicate, and all of the results are presented as means ± standard deviation (SD). A one-way ANOVA followed by the Tukey test was used to determine the significant differences, with *p*-values less than 0.05 considered significant.

## 3. Results and Discussion

### 3.1. Determination of the Total Phenolic Compounds

The application of UAE in recent years has proven to be an efficient method for the extraction of a wide spectrum of different valuable or bioactive compounds from plant materials such as carotenoids, polyphenols, polysaccharides, proteins, dietary fibers, thymols, or different oils. [13,14]. In addition, the application of UAE for the extraction of bioactive compounds from different waste and food byproducts has been successfully conducted. Moorthy et al. (2015) [15], conducted an optimization of the UAE process for the extraction of pectin from waste pomegranate peel, while Mellinas et al. (2022) [16], successfully applied UAE for the extraction of proteins and antioxidants from tomato, watermelon, and apple peels. However, one of the most important areas of UAE application is the extraction of polyphenolic compounds. A large number of studies report the application of UAE for the extraction of polyphenolic compounds from a wide range of different plant materials such as grape seeds, orange peels, olive pomace, or onion solid waste. However, it is also important to mention that although UAE can favor the extraction of phenolic compounds, the obtained extracts usually contain additional components such as organic acids, esters, or polymeric sugars [14,17–20].

Therefore, UEA was applied as a proven extraction technique for the green and efficient extraction of the total phenolic content (TPC) from the herbal dusts of green tea, hibiscus, and lemon balm, and the effects of the different extraction parameters were evaluated. All of the obtained results are expressed as gallic acid equivalents per g of dried extract (Table 3).

**Table 3.** Content of total phenolic compounds in the obtained extracts [mg GAE/g DE].

| Extract | TPC | Extract | TPC | Extract | TPC |
|---|---|---|---|---|---|
| GT 1 | 108.01 ± 0.80 $^{c}$ | HB 1 | 59.30 ± 0.51 $^{ab}$ | LB 1 | 254.09 ± 4.66 $^{d}$ |
| GT 2 | 111.89 ± 1.21 $^{c}$ | HB 2 | 60.63 ± 0.10 $^{a}$ | LB 2 | 276.90 ± 5.68 $^{c}$ |
| GT 3 | 134.33 ± 1.60 $^{b}$ | HB 3 | 59.79 ± 0.95 $^{ab}$ | LB 3 | 227.37 ± 1.25 $^{e}$ |
| GT 4 | 152.91 ± 0.74 $^{a}$ | HB 4 | 59.67 ± 0.97 $^{ab}$ | LB 4 | 356.22 ± 3.13 $^{a}$ |
| GT 5 | 75.38 ± 1.78 $^{d}$ | HB 5 | 54.51 ± 0.82 $^{c}$ | LB 5 | 278.69 ± 8.15 $^{c}$ |
| GT 6 | 77.75 ± 2.45 $^{d}$ | HB 6 | 58.32 ± 0.77 $^{b}$ | LB 6 | 319.97 ± 3.32 $^{b}$ |

Each value is the mean value of three replicates ± SD. A significant difference between samples ($p < 0.05$) is indicated by different letters within a column, for each extraction condition.

3.1.1. Total Phenolic Content—Green Tea Herbal Dust

As shown in Table 3, the TPC of the green tea extracts was in the range from 75.38 ± 1.78 to 152.91 ± 0.74 mg GAE/g DE, reaching the maximum value at the conditions of 60% amplitude and the 10 min of extraction time (GT 4). The increase in the extraction amplitude and the time of the extraction positively impacted the TPC of the green tea herbal dust extracts, up to the maximum point reached for the GT 4 extract, after which a significant reduction in the TPC was recorded (Figure 1). Similar results were recorded by Garcia-Larez et al. (2024) [21], while applying UAE for the recovery of the polyphenols from pecan nutshells, where the highest TPC was recorded for the amplitude of 50 and 60%, while the additional increase in the extraction amplitude led to a significant drop in the TPC. Wang et al. (2020) [22], also reported a similar trend in the UAE of polyphenols from potato peel. This reduction in the TPC can be attributed to the mechanisms of the UAE where the acoustic cavitation is influenced by the amplitude of the ultrasound waves. With the increase in the amplitude of the extraction, more power is delivered to the matrix, which influences the increase in the extraction efficiency due to better solvent penetration and the disruption in the molecular bonds between bioactive compounds and the matrix. However, with applying higher amplitudes, the creation of free radicals can occur, which can lead to the degradation of polyphenols, and therefore to the reduction in TPC [22–24].

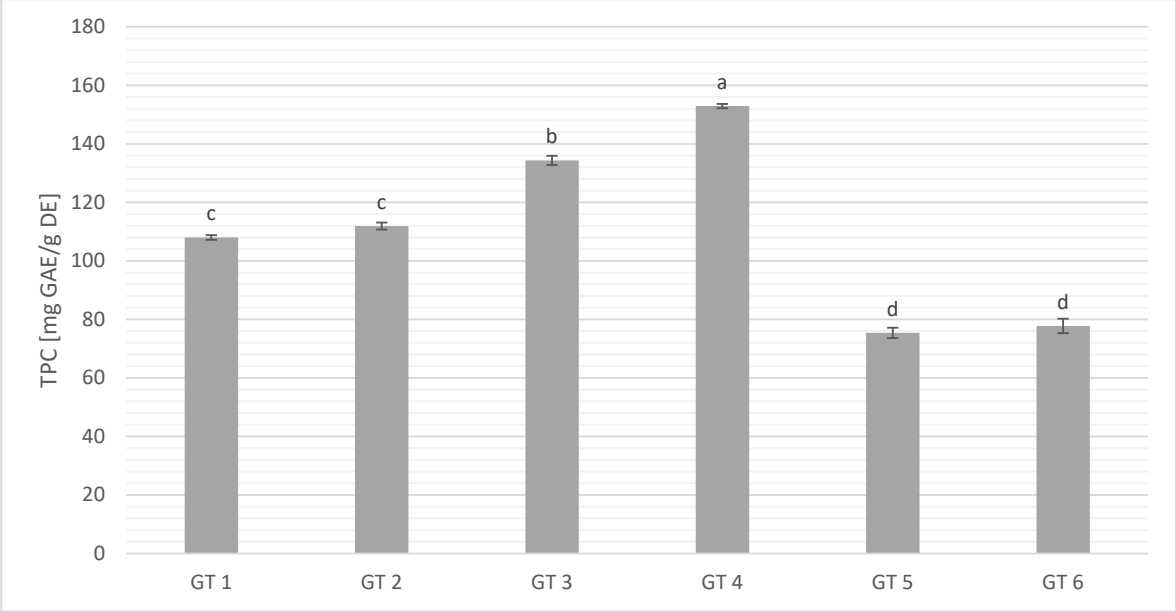


**Figure 1.** The total phenolic content obtained in extracts of green tea herbal dust (GT1–GT6); significant difference between samples ($p < 0.05$) is indicated by different letters.

In recent years, different extraction procedures for green tea have been the subject of many studies; however, there is a limited number of studies that focused on combining green extraction technologies and the utilization of green tea waste or herbal dust [25,26]. Additionally, to our knowledge, no study aimed to cover the complete utilization of green tea herbal dust. Analyzing the results from the available literature, it can be concluded that the TPC obtained from green tea herbal dust within this study is comparable to the results obtained by the traditional extraction techniques of green tea, like infusions or different variations of maceration. Barreira et al. (2021) [27], conducted four different extraction methods including hot infusion extraction for 5 to 7 min, hot infusion extraction for 30 min, maceration with water as solvent (48 h), and methanolic extraction for the commercially available green tea from the Azores, which resulted in extract TPC of 151.843, 147.850, 111.793, and 85.18 mg GAE/g DE, respectively. In addition, the same author conducted maceration for three more commercially available green tea from China, Japan, and Sri

Lanka, resulting in TPC of 65.01, 62.13, and 80.68 mg GAE/g DE, respectively [28]. In contrast, Luo et al. (2020) [29], reported that values of TPC for UAE of the green tea obtained from Hubei, China ranged between 219.00 and 243.00 mg GAE/g DE, depending on the solvent. These results are up to 37.07% or 1.58 times higher than the highest value recorded in this study, GT 4 extract (152.91 mg GAE/g DE). The lower yield of TPC within this study can be attributed to the different origins of the plant material, but it also can be the consequence of the improper storage procedure of the herbal dust, which is as previously said regarded as a waste. Therefore, this study implies that by utilization of green tea herbal dust for UAE, yields comparable to the extraction of fresh material can be provided, with the added benefit of re-valorization and further utilization of the plant material.

#### 3.1.2. Total Phenolic Content—Hibiscus Herbal Dust

TPC of the hibiscus herbal dust extracts was in the range from 58.32 ± 0.77 to 60.63 ± 0.10 mg GAE/g DE (Figure 2), with the highest concentration recorded in the extract HB 2 extract, which was obtained at the UAE conditions of 100% amplitude and 10 min of extraction time. Although the highest concentration of TPC was recorded for the HB 2 extract, the analysis showed that there is no significant difference between TPC obtained for the other extraction conditions. Similar to the results of this study, Peredo Pozos et al. (2020) [30], conducted a water bath UAE of *H. sabdariffa* with four different sonication times: 20, 40, 60, and 120 min, and reported TPC of 11.46, 12.71, 13.01, and 12.94 mg GAE/g DE, respectively. The authors reported no significant difference in the TPC with the increase in the sonication time, except with the increase from 20 to 40 min sonication. In comparison with the results of this study, UAE with the probe provided 4.66 times higher TPC than the highest TPC provided with water bath UAE. Comparing the results of TPC provided by UAE with the traditional solvent extraction techniques of *H. sabdariffa* in the study conducted by Sindi et al. (2014) [31], water, methanol, ethyl acetate, and hexane extraction, with and without the addition of formic acid, was investigated. The extracts with pure water as a solvent provided the highest TPC of 21.67 mg GAE/g, which was around 2.8 times lower than the TPC of the HB 2 extract. In addition, within the previously mentioned study published by Peredo Pozos et al. (2020) [30], solid–liquid (S-L) extraction of *H. sabdariffa* was also conducted using 80% ethanol as a solvent and for a duration of 72 h. The study reported that S-L extraction achieved the highest TPC of 65.28 mg GAE/g DE, which was slightly higher than the TPC of HB 2 extract, therefore indicating that the selection of solvent can have a great impact on the yield of total phenols.

Results obtained within this study indicate that the application of UAE is suitable for the recovery of hibiscus herbal dust phenolic compounds, while also indicating that the *H. sabdariffa* phenolic compounds are resistant to the degradation that occurs at the higher amplitudes of UAE. Additionally, the results also imply that the creation of hibiscus herbal dust positively impacts the recovery of phenolic compounds as it reduces the size of the particles and therefore increases the surface area contact between the solvent and the matrix. In accordance, the yield of the TPC obtained in this study was significantly higher than recorded yields in the available literature, where similar extraction techniques were applied.

#### 3.1.3. Total Phenolic Content—Lemon Balm Herbal Dust

The content of TPC recorded for the extracts of lemon balm herbal dust ranged between 227.37 ± 1.25 and 356.22 ± 3.13 mg GAE/g DE (Figure 3). Similar to the extraction of green tea herbal dust, the highest recorded TPC was at the UAE conditions of 60% amplitude and 10 min of extraction time (LB 4). However, in the case of lemon balm herbal dust extraction, significantly higher values were recorded for the 20% amplitude UAE, compared to the 100% amplitude for the same extraction time. In addition, no significant difference in extract TPC was recorded for the extraction conducted on the highest amplitude and longest extraction time (extract LB 2) and the extract obtained at the lowest amplitude and 5 min of extraction (extract LB 5). Therefore, these results indicate that an increase

in the power output of the UAE can lead to the significant degradation of polyphenolic compounds, which also provides insight into the physical characteristics of lemon balm bioactive compounds. As previously mentioned, several studies also reported a similar decrease in the TPC with the increase in the amplitude of UAE [21,22].

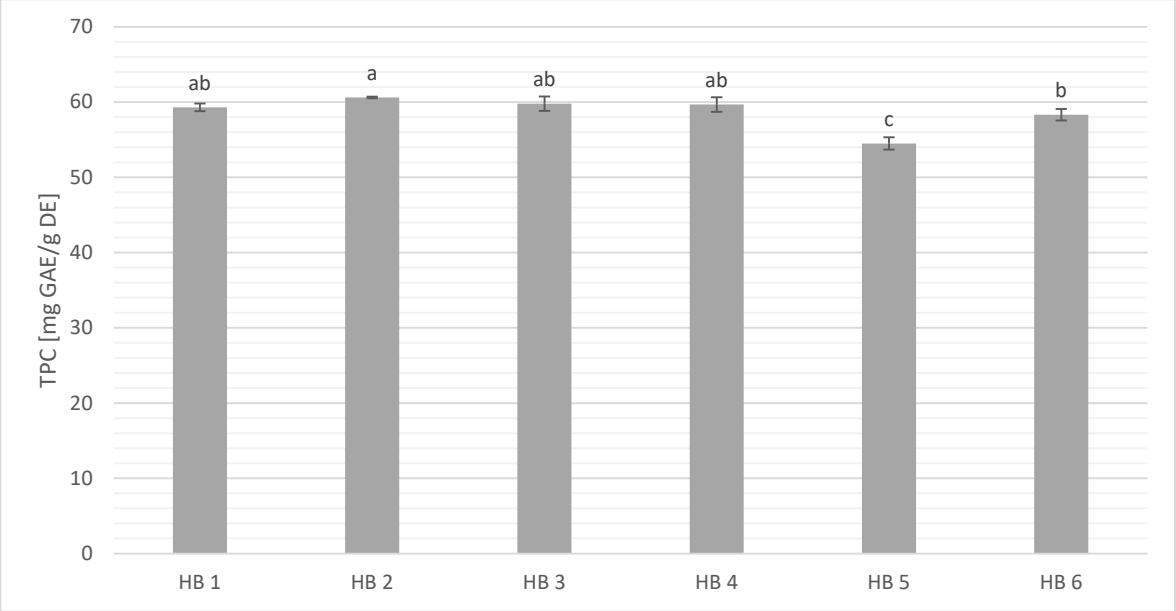


**Figure 2.** Total phenolic content obtained in extracts of hibiscus herbal dust (HB1–HB6); significant difference between samples ($p < 0.05$) is indicated by different letters.

In comparison with the available studies that described the extraction of lemon balm, the results of this study were comparable with the results obtained by traditional techniques that used water as a solvent for lemon balm extraction. In the study conducted by Petkova et al. (2017) [32], eight different commercially available lemon balm teas were extracted by hot infusion for 20 min, and the TPC was measured. Compared to the TPC of the LB 4 extract (calculated as mg GAE per g of dried plant material), no significant difference was recorded for four of the extracted samples, with only two extracted samples recording higher TPC values than the LB 4 extract. However, in the study conducted by Mabrouki et al. (2018) [33], hexane and ethanol were used for the Soxhlet extraction of lemon balm for the duration of 6 h. The recorded TPC was 1.01 mg GAE/g DE for hexane and 63.00 mg GAE/g DE for ethanol extract, which was significantly lower than the TPC recorded for all of the extracts obtained within this study, further emphasizing the polarity of polyphenols present in lemon balm. In comparison with the novel extraction technologies, Ziagova et al. (2022) [34], conducted a pulsed electric field extraction (PEFE) combined with UAE for the extraction of lemon balm, where 3.47 times higher TPC was achieved than the TPC of LB 4 extract (calculated as mg GAE/g DW). Similarly, the application of UAE and microwave-assisted extraction (MAE) for the extraction of commercially available lemon balm from Turkey achieved TPC 2.36 and 3.27 times higher than the TPC of LB 4 extract, calculated per g of dry material [35]. As previously discussed, even though the TPC of lemon balm herbal dust extracts was not as high as extracts obtained by novel extraction techniques in the above-mentioned studies, it should be kept in mind that this study utilized material that was generally considered waste. Additionally, the plant material used in this study was later converted into biochar and integrated into the final product, therefore emphasizing the value and possible utilizations of plant herbal dust material.

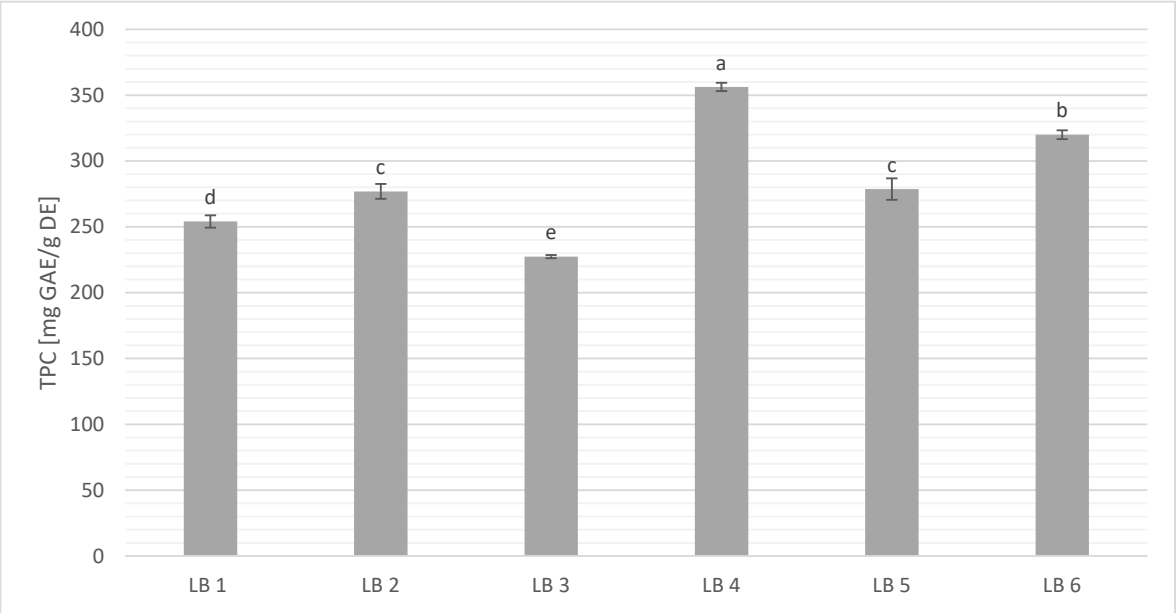


**Figure 3.** Total phenolic content obtained in extracts of lemon balm herbal dust (LB1–LB6); significant difference between samples ($p < 0.05$) is indicated by different letters.

### *3.2. HPLC Analysis of Green Tea Herbal Dust Extracts*

According to several studies published in recent years, principal phenolic components present in green tea extracts belong to the group of catechins, which mainly include non-ester catechins ((-)-epicatechin (EC) and (-)-epigallocatechin (EGC)), and ester catechins ((-)-epicatechin gallate (EGC) and (-)-epigallocatechin gallate (EGCG)). In addition, epicatechins can change to their epimer forms, which are known as (+)-catechin (C), (-)-gallocatechin (GC), and (-)-gallocatechin gallate (GCG) [36,37]. Considered the main carriers of the health-beneficial activity of green tea extracts, several studies report that catechins exhibit hepatoprotective, cardioprotective, anti-obesity, antibacterial, antioxidant, and antiviral activity [38–40]. Among the green tea catechins, EGCG is generally considered to possess the highest activity and is therefore researched the most, mainly in the field of antioxidant and anticancer activity [41]. Focused on the EGCG anticancer activity, comprehensive research conducted by Singh et al. (2011) [42], reported results of several clinical studies, which indicated preventive, proapoptotic, and antiproliferative effects.

In this study, HPLC analysis was conducted for all of the obtained green tea herbal dust extracts, and the presence of four characteristic components was recorded (Figure 4). As demonstrated in Table 4, the green tea herbal dust extracts contained gallic acid (GA) (43.38 ± 0.11–101.12 ± 0.18 mg/L), EGC (314.94 ± 0.76–703.51 ± 3.30 mg/L), EGCG (503.35 ± 1.99–1296.94 ± 9.56 mg/L), and GCG (30.3 ± 0.14–75.02 ± 0.25 mg/L). The most dominant component in extracts obtained on all of the UAE conditions was EGCG, recording the maximum concentration in extract GT 4 (1296.94 ± 9.56 mg/L). In addition, the results obtained by HPLC analysis were in correlation with the TPC of the extracts; therefore, the concentration of bioactive compounds increased with the increase in the amplitude of the UAE. Accordingly, significantly higher concentrations of GA, EGC, EGCG, and GCG were recorded for the GT 4 extract, while the 20% amplitude of UAE exhibited the lowest yields.

In comparison with the available literature, Hu et al. (2009) [43], reported a study where an orthogonal experimental design was used for the optimization of ethanol-based solvent green tea extraction and the creation of EGC- and EGCG-enriched green tea extracts. The authors reported that the highest achieved contents of EGC and EGCG were 195.9 and 282.8 mg/L, respectively, which is significantly lower than the recorded EGC and EGCG contents achieved by UAE used in this study, even on the 20% amplitude. In the study

conducted by Luo et al. (2020) [29], the yield of EGC (22.00–24.40 mg/g DE) and EGCG (36.80–94.00 mg/g DE) was achieved using four different types of green tea extraction (UAE-DES, UAE–ethanol, ethanol–solvent extraction, and hot infusion). Compared with the results of this study, calculated per g of DE, the yield of EGC was in the range of 45.30 to 49.83 mg/g DE, and the yield of EGCG was in the range of 70.87 to 90.41 mg/g DE. Therefore, the application of UAE for the extraction of green tea herbal dust achieved higher concentrations of EGC and comparable yields of EGCG as the extraction of commercially available green tea. Additionally, Ayyildiz et al. (2018) [36], conducted an optimization of EGCG extraction from commercially available green tea in Turkey, applying hot infusion and UAE with water as a solvent, and UAE with ethanol as a solvent. They reported the EGCG yields of 38.6 ± 0.02, 48.1 ± 0.04, and 76.2 ± 0.03 mg/g DW, respectively, which was significantly lower than the EGCG concentration obtained in the GT 4 extract. Considering the above-mentioned, it can be concluded that UAE can be successfully applied for the recovery of green tea herbal dust bioactive components while creating the additional possibility of further utilization of plant material by turning it into usable bio-char. In addition, HPLC analysis showed that the obtained components possess strong health-beneficial properties, creating the potential for further analysis and research focusing on the application of green tea herbal dust extracts.

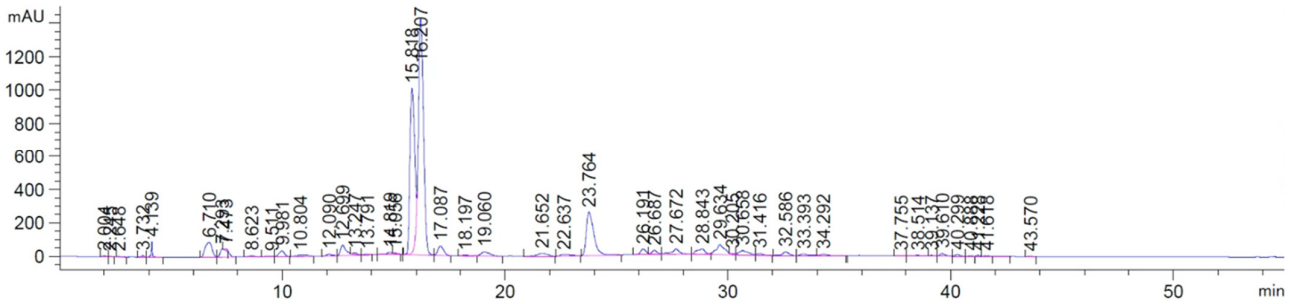


**Figure 4.** Chromatogram of green tea herbal dust extract HPLC analysis.

**Table 4.** The concentration of phenolic compounds in extracts of green tea herbal dust obtained by UAE expressed as mg/L.

| Extract | Gallic Acid | Epigallocatechin (EGC) | Epigallocatechin Gallate (EGCG) | Gallocatechin Gallate (GCG) |
|---|---|---|---|---|
| GT 1 | 95.96 ± 0.81 $^{c}$ | 637.88 ± 27.71 $^{c}$ | 997.86 ± 12.61 $^{c}$ | 68.08 ± 0.23 $^{c}$ |
| GT 2 | 100.39 ± 0.33 $^{a}$ | 684.46 ± 1.42 $^{ab}$ | 1121.54 ± 6.30 $^{b}$ | 71.23 ± 0.01 $^{b}$ |
| GT 3 | 98.77 ± 0.40 $^{b}$ | 694.71 ± 4.77 $^{ab}$ | 1273.02 ± 4.33 $^{a}$ | 71.06 ± 0.71 $^{b}$ |
| GT 4 | 101.12 ± 0.18 $^{a}$ | 703.51 ± 3.30 $^{a}$ | 1296.94 ± 9.56 $^{a}$ | 75.02 ± 0.25 $^{a}$ |
| GT 5 | 43.38 ± 0.11 $^{e}$ | 314.94 ± 0.76 $^{e}$ | 503.35 ± 1.99 $^{e}$ | 30.3 ± 0.14 $^{e}$ |
| GT 6 | 56.45 ± 0.06 $^{d}$ | 422.11 ± 0.13 $^{d}$ | 650.49 ± 2.83 $^{d}$ | 40.17 ± 0.26 $^{d}$ |

A significant difference between samples for each extraction condition ($p < 0.05$) is indicated by different letters within a column.

### *3.3. HPLC Analysis of Hibiscus Herbal Dust Extracts*

The HPLC analysis was conducted for all of the obtained hibiscus herbal dust extracts, and a total of nine phenolic compounds were detected, mostly belonging to the group of phenolic acids, while the presence of only three flavonoids was detected (Tables 5 and 6). The most dominant components were neochlorogenic acid (40.89 ± 10–45.70 ± 0.23 mg/L), protocatechuic acid (35.52 ± 0.07–42.05 ± 0.19 mg/L), quercetin-3-β-D-glucoside (10.11 ± 0.21–13.405 ± 0.12 mg/L), and gallic acid (8.17 ± 0.01–10.58 ± 0.40 mg/L). The content of these principal components, however, was not directly correlated with the TPC exhibited by the hibiscus herbal dust extracts, meaning that significantly higher concen-

trations of all bioactive components, except the quercetin 3-β-D-glucoside, were recorded for the HB 4 extract rather than the HB 2 extract, which achieved highest TPC. The reason behind this discrepancy in the results could have been because of the presence of specific anthocyanins that are part of the *H. sabdariffa* extracts and were not covered by standard HPLC polyphenolic analysis (Figure 5). Additionally, by analyzing the results, it can be concluded that higher concentrations of phenolic acids were achieved on UAE amplitudes of 60 and 100%, while the concentration of flavonoids was significantly higher on the UAE amplitude of 20%. These results indicated that flavonoids present in the hibiscus herbal dust extracts were significantly more susceptible to degradation influenced by higher UAE power outputs, which is also reported by other authors for other plant materials [21]. Comparing the results obtained within this study with the available literature, Ifie et al. (2016) [44], conducted a solvent extraction using distilled water at the temperature of 50 °C for the extraction of three different varieties of *H. sabdariffa* dried calyxes (dark red, light red, and white), and the obtained extracts were analyzed by HPLC. The authors reported a total of 24 phenolic components, including all of the components detected within this study. As previously mentioned, the anthocyanins were the most dominant phenolic compounds in the obtained extract, except for the white variety of *H. sabdariffa* calyxes, while the content of phenolic acids and flavonoids was comparable to the results of this study, calculated per g of dried weight (DW).

Regarding the pharmaceutical activity of the obtained components in hibiscus herbal dust extracts, in recent years, many authors reported studies investigating their health-beneficial properties. Therefore, a review conducted by Kahkeshani et al. (2019) [45], reported that gallic acid exhibits antimicrobial and anticancer activity, in addition to beneficial effects in treatments of gastrointestinal, cardiovascular, metabolic, and neuropsychological diseases. Antimicrobial, anticancer, and activity against cardiovascular diseases were also reported activities for both quercetin and myricetin [46,47], while Kim et al. (2016) [48], reported that quercetin 3-β-D-glucoside improved memory and cognitive dysfunction and also exhibited neuroprotective activity. As a type of naturally occurring and widely distributed phenolic acid, protocatechuic acid was comprehensively researched for its many biological activities. A review study conducted by Kakkar and Bais, (2014) [49], reports that protocatechuic acid possesses antibacterial, antioxidant, neuroprotective, chemoprotective, and anti-inflammatory activity. In conclusion, the hibiscus herbal dust extracts obtained by UAE exhibited concentrations of bioactive components comparable to the results in the available studies, although a more detailed analysis would be required to have a complete phenolic profile of these extracts. However, the components that were detected exhibit proven health-beneficial activities, therefore making these extracts a potentially valuable pharmaceutical product.

**Table 5.** The concentration of phenolic compounds in extracts of hibiscus herbal dust obtained by UAE expressed as mg/L.

| Extract | Gallic Acid | Protocatechuic Acid | Neochlorogenic Acid | Caffeic Acid | p-Coumaric Acid |
|---|---|---|---|---|---|
| HB 1 | $8.17 \pm 0.01^{d}$ | $35.52 \pm 0.07^{e}$ | $40.89 \pm 0.10^{e}$ | $2.31 \pm 0.01^{d}$ | $4.23 \pm 0.05^{bcd}$ |
| HB 2 | $9.14 \pm 0.03^{cd}$ | $37.82 \pm 0.16^{d}$ | $43.25 \pm 0.38^{bc}$ | $2.40 \pm 0.01^{c}$ | $4.13 \pm 0.25^{d}$ |
| HB 3 | $9.64 \pm 0.48^{abc}$ | $39.96 \pm 0.11^{b}$ | $43.64 \pm 0.02^{b}$ | $2.57 \pm 0.02^{ab}$ | $4.50 \pm 0.04^{abc}$ |
| HB 4 | $10.58 \pm 0.40^{a}$ | $42.05 \pm 0.19^{a}$ | $45.70 \pm 0.23^{a}$ | $2.60 \pm 0.01^{ab}$ | $4.66 \pm 0.04^{ab}$ |
| HB 5 | $10.25 \pm 0.01^{ab}$ | $38.80 \pm 0.01^{c}$ | $42.40 \pm 0.05^{cd}$ | $2.63 \pm 0.01^{a}$ | $4.71 \pm 0.02^{a}$ |
| HB 6 | $9.30 \pm 0.06^{bc}$ | $38.03 \pm 0.28^{d}$ | $41.91 \pm 0.34^{d}$ | $2.35 \pm 0.03^{cd}$ | $4.69 \pm 0.08^{a}$ |

A significant difference between samples for each extraction condition ($p < 0.05$) is indicated by different letters within a column.

**Table 6.** The concentration of phenolic compounds in extracts of hibiscus herbal dust obtained by UAE expressed as mg/L.

| Extract | Trans-Ferulic Acid | Quercetin 3-β-D-Glucoside | Myricetin | Quercetin |
|---|---|---|---|---|
| HB 1 | 3.38 ± 0.11 $^{b}$ | 11.26 ± 1.08 $^{a}$ | 3.94 ± 00 $^{d}$ | 3.92 ± 0.05 $^{b}$ |
| HB 2 | 3.55 ± 0.08 $^{b}$ | 11.06 ± 2.12 $^{a}$ | 3.96 ± 0.01 $^{cd}$ | 3.50 ± 0.28 $^{b}$ |
| HB 3 | 3.74 ± 0.21 $^{ab}$ | 11.05 ± 0.04 $^{a}$ | 3.97 ± 00 $^{bc}$ | 5.31 ± 0.38 $^{a}$ |
| HB 4 | 4.04 ± 0.09 $^{a}$ | 13.41 ± 0.12 $^{a}$ | 3.97 ± 0.01 $^{b}$ | 5.57 ± 00 $^{a}$ |
| HB 5 | 3.72 ± 0.06 $^{ab}$ | 13.52 ± 0.03 $^{a}$ | 4.28 ± 0.01 $^{a}$ | 5.72 ± 0.01 $^{a}$ |
| HB 6 | 3.63 ± 0.09 $^{ab}$ | 10.11 ± 0.21 $^{a}$ | 3.97 ± 00 $^{bc}$ | 5.20 ± 0.25 $^{a}$ |

A significant difference between samples for each extraction condition ($p < 0.05$) is indicated by different letters within a column.

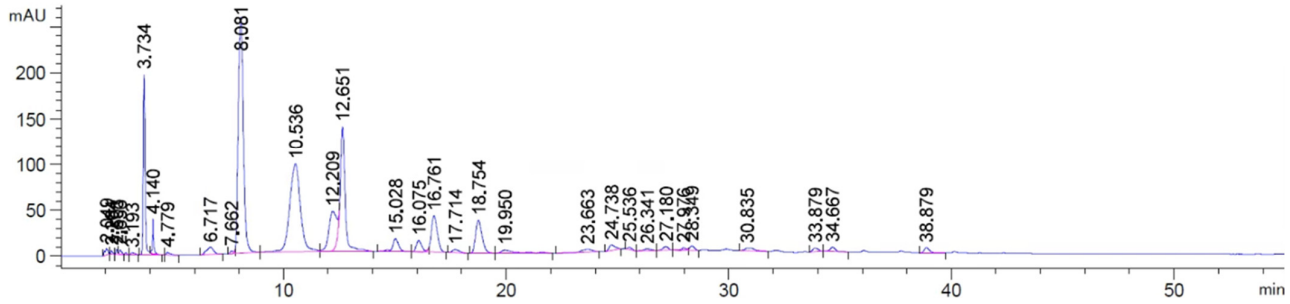


**Figure 5.** Chromatogram of hibiscus herbal dust extract HPLC analysis.

### *3.4. HPLC Analysis of Lemon Balm Herbal Dust Extracts*

As for the green tea and hibiscus herbal dust extracts, HPLC analysis was also conducted for all of the obtained lemon balm herbal dust extracts, and the presence of four characteristic phenolic compounds was confirmed (Table 7). The protocatechuic acid (0.89 ± 00–1.89 ± 0.01 mg/L), caffeic acid (7.18 ± 0.05–13.41 ± 0.01 mg/L), rutin (5.79 ± 0.04–12.85 ± 0.02 mg/L), and rosmarinic acid (220.26 ± 0.95–521.39 ± 2.38 mg/L) were detected (Figure 6), with the rosmarinic acid achieving the highest concentration in all of the obtained extracts. These results are in accordance with a number of studies reporting that rosmarinic acid is the principal component of lemon balm extracts [50–52]. In addition, the concentrations of detected bioactive components, calculated as mg per g DE, were in correlation with the TPC, meaning that the highest values were obtained for the extract LB 4. Accordingly, calculated per g of DE, the UAE on the 20% amplitude also achieved higher concentrations of all detected compounds than the UAE on the 100% amplitude.

**Table 7.** The concentration of phenolic compounds in extracts of lemon balm herbal dust obtained by UAE expressed as mg/L.

| Extract | Protocatechuic Acid | Caffeic Acid | Rutin | Rosmarinic Acid |
|---|---|---|---|---|
| LB 1 | 1.66 ± 00 $^{b}$ | 9.71 ± 0.24 $^{c}$ | 10.61 ± 0.09 $^{b}$ | 406.00 ± 3.07 $^{b}$ |
| LB 2 | 1.89 ± 0.01 $^{a}$ | 13.41 ± 0.01 $^{a}$ | 12.85 ± 0.02 $^{a}$ | 521.39 ± 2.38 $^{a}$ |
| LB 3 | 1.21 ± 0.01 $^{e}$ | 7.18 ± 0.05 $^{e}$ | 6.95 ± 0.04 $^{e}$ | 292.06 ± 1.03 $^{e}$ |
| LB 4 | 1.49 ± 0.01 $^{c}$ | 11.76 ± 0.02 $^{b}$ | 9.00 ± 0.04 $^{c}$ | 396.11 ± 2.80 $^{c}$ |
| LB 5 | 0.89 ± 00 $^{f}$ | 8.14 ± 0.01 $^{d}$ | 5.79 ± 0.04 $^{f}$ | 220.26 ± 0.95 $^{f}$ |
| LB 6 | 1.28 ± 00 $^{d}$ | 11.83 ± 0.02 $^{b}$ | 7.70 ± 0.04 $^{d}$ | 318.29 ± 0.86 $^{d}$ |

A significant difference between samples for each extraction condition ($p < 0.05$) is indicated by different letters within a column.

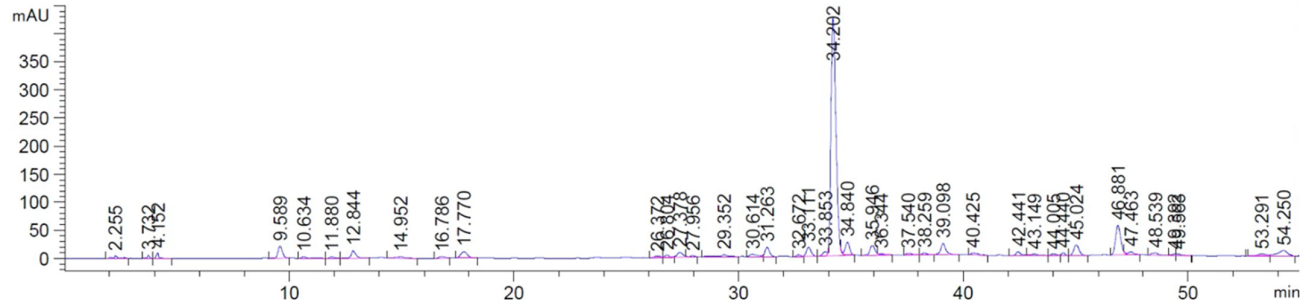


**Figure 6.** Chromatogram of lemon balm herbal dust extract HPLC analysis.

Although the HPLC analysis detected the presence of only four phenolic compounds, it is considered that caffeic acid, rutin, and rosmarinic acid are one of the main phenolic bioactive components of lemon balm extracts [8]. Magnani et al. (2014) [52], reported that caffeic acid, being widely distributed in plant food sources, possesses in vitro antioxidant and antibacterial activity, while also exhibiting cardiovascular activity and inhibitory activity on cancer cell growth. One of the most widely researched phenolic compounds, rutin, was the subject of many studies because of its wide health-beneficial activities. Ganeshpurkar and Saluja, (2017) [53], published a review paper focusing on the rutin pharmacological properties recorded in several studies, and reported that rutin exhibited effects on the nervous, cardiovascular, gastrointestinal, and respiratory systems, with an emphasis on anticancer, hepatoprotective, and nephroprotective activity. The most abundant compound in lemon balm extracts, and the ester of caffeic acid, rosmarinic acid is distributed among a number of plants, mostly from the Lamiaceae family. Due to the large number of pharmacological activities, rosmarinic acid is widely researched and many studies reported on its health-beneficial activities. A comprehensive review by Nadeem et al. (2019) [54], reported that rosmarinic acid exhibits anticancer, antidiabetic, antimicrobial, and cardioprotective activity, while also showing antidepressant, neuroprotective, and antioxidant effects.

In comparison with the available literature, Petrisor et al. (2022) [8], published a review paper where the results of several studies on the subject of *Mellisa officinalis* polyphenol extractions were reported. The authors reported that major phenolic compounds included a total of eight different phenolic acids and 11 different flavonoids, with rosmarinic acid, rutin, and caffeic acid being the most dominant. Ghiulai et al. (2020) [55], reported the results of the freshly harvested and air-dried lemon balm extraction using two different maceration extraction and sonication extraction with 70% ethanol, 96% ethanol, and 80% methanol as a solvent, respectively. The study reported that the concentration of the rosmarinic acid was in the range from 8.63 to 86.63 mg/g DW, where the highest value was comparable with the concentration obtained in extract LB 4, calculated per g of DW. Additionally, Ghiulai et al. (2020) [55], also reported concentrations of caffeic acid and rutin (0.86 and 0.85 mg/g DW, respectively), which were significantly lower than the concentrations of caffeic acid and rutin obtained within this study. Similar results were also reported by Silva et al. (2023) [56], where commercially available lemon balm was extracted by applying the infusion, dynamic maceration, and decoction, and achieved a yield of rosmarinic acid of 34.41, 40.36, and 41.71 mg/g DE, respectively, which was significantly lower than the recorded values within this study. Considering the results reported by the above-mentioned studies, it can be concluded that the UAE of the lemon balm herbal dust provided the extracts with major bioactive phenolic compounds in concentrations higher or comparable with the traditional, widely used extraction techniques.

## 4. Conclusions

This study aimed to assess the quality of the green tea, hibiscus, and lemon balm herbal dust extracts obtained by UAE in terms of TPC and phenolic profile. Herbal dust or dried and grounded plant material with particle sizes lower than 0.315 mm, which is

usually considered waste by the tea industry, was extracted by UAE, and the obtained results were compared with the available literature. In addition, different conditions of UAE were investigated in order to maximize the yield of the extraction, and therefore increase the concentration of the bioactive compounds. The results indicated that the UAE of green tea and lemon balm herbal dust on 60% amplitude, and UAE of hibiscus herbal dust on 100% amplitude, for 10 min, provided extracts with TPC comparable with the traditional extraction techniques as well as several novel extraction methods of commercially available plant materials. The phenolic profile analyzed by HPLC analysis also showed comparable or even higher concentrations of principal bioactive phenolic compounds than the extracts of commercial plant material. In addition, the HPLC analysis of the obtained extracts showed the presence of compounds with proven pharmaceutical activity, notably anticancer, cardiovascular, neuroprotective, or hepatoprotective. The highest concentration of bioactive compounds exhibited by herbal dust extracts was EGCG for green tea (1296.94 $\pm$ 9.56 mg/L), neochlorogenic acid for hibiscus (45.70 $\pm$ 0.23 mg/L), and rosmarinic acid for lemon balm (521.39 $\pm$ 2.38 mg/L). Considering the above-mentioned, it can be concluded that the UAE can be successfully applied for the extraction of green tea, hibiscus, and lemon balm herbal dust, and provided extracts rich in bioactive compounds, comparable with the extracts of commercial plant materials. Additionally, the cellulosic material left over from the extraction procedure will be used for the subsequent study, where it will be incinerated and integrated into rubber as a filler compound; therefore, the herbal dust will be completely utilized without the creation of additional waste in the process.

**Author Contributions:** Conceptualization, S.S. and J.L.S.; methodology, S.S., K.A. and S.J.; software, S.S. and K.A.; validation, A.G. and S.V.; formal analysis, S.S., K.F. and K.A.; investigation, S.S., K.F. and K.A.; resources, S.S., K.A. and S.J.; data curation, S.S. and K.F.; writing—original draft preparation, S.S. and K.F.; writing—review and editing, A.G. and S.V.; visualization, S.S. and K.F.; supervision, A.G. and S.V. All authors have read and agreed to the published version of the manuscript.

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