

## Article

# Nutraceutical Value of Eleven Aromatic Medicinal Plants and Azorean *Camellia sinensis*: Comparison of Antioxidant Properties and Phenolic and Flavonoid Contents

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**Abstract:** Drug discovery based on medicinal plants remains an important source of bioactive compounds, many of which have been the basis for new chemical structures for the pharmaceutical and food industries. According to the World Health Organization, about 80% of the worldwide population still depends on plant drugs, and several medicines have been obtained from medicinal plants. Unfortunately, the potential benefits of these plants have led to unscientific exploration of natural resources, a fact that is being globally observed. The aim of this study was to evaluate eleven aromatic medicinal plants and compare them to Azorean *Camellia sinensis* green tea in terms of antioxidant activity, total phenolics, and flavonoid content, and also to evaluate the possibility of their valorization as a nutraceutical material. The results revealed that *Camellia sinensis* presented higher values for free radical scavenging activity (FRSA, EC<sub>50</sub> = 3.43 µg/mL), ferric reducing antioxidant power (FRAP, EC<sub>50</sub> = 5.12 µg/mL), and total phenolic content (TPC, 294.43 mg acid gallic equivalents per g of dry extract (DE)). However, the aromatic medicinal plants also presented significant results in terms of FRSA and FRAP, particularly *Aloysia citrodora*, *Mentha pulegium*, and *Stevia rebaudiana*. For ferric ion chelating (FIC), the highest value was found in *Cymbopogon citratus* (80.60%). *Mentha pulegium* and *Aloysia citrodora* had significant values for TPC (199.15 and 187.15 mg GAE/g DE, respectively), but were lower than the values of *Camellia sinensis*. For flavonoid content (TFC), the highest value was shown in *Achyrocline satureioides* (265.75 mg rutin equivalents per g DE). This study revealed the importance of some aromatic medicinal plants in terms of bioactivities, and that their combination with green tea is a perfect blend for added value with beneficial nutraceutical effects for human health.

**Keywords:** medicinal herbs; *Camellia sinensis*; polyphenolic compounds; antioxidant activities; therapeutic effects



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## 1. Introduction

Natural bioactive compounds have long been used as important sources for therapeutic drugs and biodiversity and have significantly contributed to human livelihood, and thus play a predominant role in the well-being of the global population. Additionally, aromatic medicinal plants have been used worldwide, particularly in the Middle East, since approximately 5000 BC for their biological properties as food preservatives and for enhancing the aroma and flavor of foodstuff [1]. Medicinal plant-based drugs have been used as traditional medicines to ameliorate several diseases due to their calming effects and antioxidant, anti-inflammatory, antimicrobial, and antiseptic properties, among other properties, and they may also reduce the risk of cancer and/or cardiovascular diseases [2]. These potential therapeutic effects are due to many active phytochemicals such as flavonoids, polyphenols, terpenoids, carotenoids, curcumins, and vitamins. Generally,

the bioactive compounds in aromatic medicinal plants play a significant role in oxidative stress protection because of their ability to protect the body from damage caused by free radicals [2,3]. Natural antioxidants from plants are classified into three major classes: phenolic compounds, carotenoids, and vitamins. Phenolic compounds show a large diversity of structures, from simple molecules (e.g., gallic acid, vanillic acid, ferulic acid, and caffeic acid) to polyphenols like flavonoids and tannins [4]. In food technology, antioxidants are frequently added to many foodstuffs in order to decrease or inhibit the occurrence of oxidation and to enrich the foods with preservative substances, thereby increasing the food's shelf life. One example is the study by Lu et al. [5] that used green tea extracts to improve the quality of sponge cake (addition of 20% of green tea extracts instead of wheat flour presented a much better pleasant taste).

An estimation from the World Health Organization (WHO) reported that 88% of all countries still use traditional medicines, such as herbal medicines, indigenous therapies, acupuncture, yoga, homeopathy, traditional Chinese medicine, naturopathy, and others. According to some authors [6], the population in developing countries still relies on traditional plants for maintaining and improving health conditions. All plants produce phytochemical substances and compounds that have pharmacological/therapeutic activities that can be used for applications in modern medicine [7]. Unfortunately, the decline in biodiversity, mainly due to the global population increasing, as well as indiscriminate deforestation, pollution, global climate changes, and overexploitation of natural resources, have had a strong impact on the preservation of aromatic medicinal plants. Therefore, it is important to preserve plant biodiversity to maintain future structural diversity for the sustainable advance of human civilization.

*Camellia sinensis* is also a plant with scientifically recognized beneficial effects on human health, and a blend with aromatic medicinal plants can add a surplus value for conscious consumers. This study evaluates eleven aromatic plants: *Aloysia citrodora*, *Cymbopogon citratus*, *Mentha pulegium*, *Pimpinella anisum*, *Stevia rebaudiana*, *Lavandula angustifolia*, *Thymus citriodorus*, *Achyrocline satureioides*, *Salvia rosmarinus*, *Laurus nobilis*, and *Hibiscus rosa-sinensis*. As described in Table 1, several aromatic medicinal plants have a variety of benefits, including pharmacological, pharmaceutical, and cosmetic applications.

**Table 1.** Local name, geographic distribution, and traditional applications of the aromatic medicinal plants.

Species	Local Name	Geographical Distribution	Traditional Applications	References
<i>Aloysia citrodora</i>	Lemon verbena	South America, North Africa, and Southern Europe	Anxiety; insomnia; gastrointestinal, respiratory, and cardiovascular problems; and rheumatism.	[8]
<i>Cymbopogon citratus</i>	Lemon grass	Asia, Africa, and South and North America	Anti-fungal, anti-bacterial, antiprotozoal, antioxidant, anti-inflammatory, anti-carcinogenic, anti-rheumatic, and cardio-protective activities.	[9,10]
<i>Mentha pulegium</i>	Pennyroyal	Europe, North Africa, and the Middle East	Colds, influenza, abdominal cramps, tuberculosis and smallpox, and latent menstruation promotion.	[11]
<i>Pimpinella anisum</i>	Anise	Mediterranean region	Diuretic and carminative, useful for epilepsy and melancholy. Used in cosmetics and perfumes.	[12]
<i>Stevia rebaudiana</i>	Honey leaf, or Sweet-leaf	South America, particularly Brazil and Paraguay	Anti-diabetic, anti-hypertensive, anti-tumor, anti-cariogenic, anti-inflammatory, and bactericidal effects.	[13]

Table 1. Cont.

Species	Local Name	Geographical Distribution	Traditional Applications	References
<i>Lavandula angustifolia</i>	Lavanda	Southern Europe and the Mediterranean area	Anti-inflammatory, anxiolytic, antidepressant, sedative, and neuroprotective properties. Used in cosmetics and perfumes.	[14]
<i>Thymus citriodorus</i>	Lemon thyme	Southern Europe, cultivated in the Mediterranean area	Cytoprotective, antioxidant, and anti-bacterial. Used for culinary purposes.	[15]
<i>Achyrocline satureioides</i>	Marcela	Brazil, Argentina, Uruguay, and Paraguay	Digestive ailments, anti-inflammatory, anti-atherosclerotic, sedative, neuroprotective, and antioxidant.	[16]
<i>Salvia rosmarinus</i>	Rosemary	Mediterranean region	Antioxidant and anti-inflammatory properties. Used as a spice for cooking.	[17]
<i>Laurus nobilis</i>	Laurel	Europe	Antioxidant, anti-inflammatory, anti-viral, anti-bacterial, anti-fungal, and immunomodulatory activity. Used in the culinary and food industries as a flavoring agent.	[18]
<i>Hibiscus rosa-sinensis</i>	Hibiscus	China and tropical regions	Antioxidant, analgesic, anti-cancer, and anti-fungal properties.	[19]

Taking into account the considerable importance of aromatic medicinal plants in the daily life of modern society and their association with health benefits, the main objective of this study was to compare the antioxidant capacity and phenolic and flavonoid contents of the eleven aromatic medicinal plants referred to above and to compare their properties with Azorean *Camellia sinensis* green tea, considered as a plant with the highest antioxidant properties.

## 2. Material and Methods

### 2.1. Chemicals and Reagents

Gallic acid (98%–G7384), rutin, (2,2-Diphenyl-1-(2,4,6-trinitrophenyl) hydrazyl (DPPH) ethylenediaminetetraacetic disodium salt (EDTA), Folin–Ciocalteu reagent (FCR), aluminum chloride (AlCl<sub>3</sub>), potassium ferricyanide, iron (II) chloride (FeCl<sub>2</sub>), iron (III) chloride (FeCl<sub>3</sub>), ferrozine, and trichloroacetic acid (TCA) were all obtained from Sigma–Aldrich (St. Louis, MO, USA). Ascorbic acid, sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>), potassium acetate (KCH<sub>3</sub>CO<sub>2</sub>), sodium phosphate, and sodium chloride (NaCl) were obtained from E. Merck (Darmstadt, Hessen, Germany). Ultrapure distilled water that was deionized with the Millipore Milli-Q purification system (Millipore, Bedford, MA, USA) was used throughout all the experiments.

### 2.2. Preparation of Aromatic Medicinal Plants and Tea Samples

The eleven aromatic plants (*Aloysia citrodora*, *Cymbopogon citratos*, *Mentha pulegium*, *Pimpinella anisum*, *Stevia rebaudiana*, *Lavandula angustifolia*, *Thymus citriodorus*, *Achyrocline satureioides*, *Salvia rosmarinus*, *Laurus nobilis*, and *Hibiscus rosa-sinensis*) were purchased from a marketplace, and Azorean *Camellia sinensis* green tea was donated by the Gorreana Tea Plantation (São Miguel Island, Azores, Portugal—37°48'57" N, 25°24'9" W). Knowing the effect of sample preparation conditions (e.g., solvent type, temperature of extraction, contact time, particle size, and solute/solvent ratio) on the preservation of antioxidant activity, the extraction yield was performed using an aqueous solution to mimic the tea infusions used by the general population. The extraction procedure was carried out using the following conditions: A dried powder material (1 g) was added to 20 mL of distilled

water and heated at 70 °C for 15 min to avoid the degradation of compounds that can occur at temperatures higher than 70 °C. This process was repeated three times, and the combined extract was filtered through a cellulose acetate membrane (porosity of 0.45 µm) to remove particulate matter. The extract solution was lyophilized for further analysis.

### 2.3. Yield Determination of Aromatic Medicinal Plants and *Camellia sinensis* Green Tea Extracts

The yield of the aromatic medicinal plants and *Camellia sinensis* was calculated using the following equation: Yield (%) =  $100 \times (A - B)/W$ , where A = weight of the flask containing the extract after lyophilization, B = weight of the dry empty flask, W = weight of dry sample.

### 2.4. Determination of the In Vitro Antioxidant Activity

To determine the antioxidant properties of the bioactive natural compounds, the samples under study were assessed using different in vitro antioxidant assays, such as the DPPH free radical scavenging activity (FRSA), ferric reducing activity power (FRAP), and ferrous ion-chelating (FIC) activity.

#### 2.4.1. Determination of DPPH Free Radical Scavenging Activity (FRSA)

Free radical scavenging activity of the aromatic medicinal plants and *Camellia sinensis* extract was measured by (2,2-Diphenyl-1-(2,4,6-trinitrophenyl) hydrazyl (DPPH) based on both the electron transfer and hydrogen atom transfer reactions, according to the method of Tong et al. [20] with slight modifications [21]. In DPPH, a stable free radical reacts with the proton-donating scavenging activity of the antioxidant compounds, changing the color of the reagent's mixture from purple to bright yellow, and the color intensity can be monitored spectrophotometrically. An aliquot of 250 µL of each extract sample (mg/mL) or ascorbic acid was added to 500 µL of DPPH solution (100 µM). Ascorbic acid was used as the reference sample, and a mixture without a sample or ascorbic acid was used as the control. The Abs was measured after incubation (at room temperature in dark) at 517 nm for a period of 30 min. The FRSA was calculated as a percentage of DPPH discoloration using the following equation: FRSA (%) =  $(1 - \text{Abs}_{\text{sample}}/\text{Abs}_{\text{control}}) \times 100$ .

The results were expressed as an EC<sub>50</sub> value (µg/mL), which is defined as the sample concentration that can quench 50% of the DPPH free radicals. A lower EC<sub>50</sub> value was indicative of a higher antioxidant activity.

#### 2.4.2. Determination of Ferric Reducing Antioxidant Power (FRAP)

The FRAP was determined according to the method of Oktay et al. [22] with some modifications [21]. The FRAP of each freeze-dried extract at various concentrations was evaluated based on their abilities to reduce ferric iron (Fe<sup>3+</sup> complex to Fe<sup>2+</sup>).

Briefly, an aliquot of 400 µL of each extract sample (mg/mL) was mixed with 400 µL of phosphate buffer (200 mM, pH 6.6) plus 400 µL of potassium ferricyanide (1%, w/v), and was incubated for 20 min at 50 °C. After this period, 400 µL of TCA (10%, w/v) was added, and the mixture was centrifuged at 4000 × g for 10 min. One milliliter of the upper layer was mixed with same volume of deionized water plus 200 µL of FeCl<sub>3</sub> (0.1% w/v). Ascorbic acid was used as the reference sample. The results were expressed as the EC<sub>50</sub> value (µg/mL), which is the concentration at which the Abs was 0.5 for reducing power, and were obtained by interpolation from a linear regression analysis of concentration versus Abs at 700 nm against a blank. A lower EC<sub>50</sub> value represented a higher antioxidant activity.

#### 2.4.3. Determination of Ferrous Ion-Chelating (FIC) Activity

The FIC activity assay was also carried out to better characterize the antioxidant abilities of the studied samples, since metal chelating capacity is reported to be one of the most important mechanisms underpinning antioxidant activity [23]. FIC activity was determined according to the method of Wang et al. [23] with some modifications [21]. The chelating ability of each extract was determined by measuring the inhibition of the

Fe<sup>2+</sup>–ferrozine complex formation. An aliquot of 200 µL of each extract sample (mg/mL) was mixed with 270 µL of methanol, 10 µL of FeCl<sub>2</sub> (2 mM), and 20 µL of ferrozine (5 mM). After 10 min at room temperature, the Abs (562 nm) was measured. Methanol, instead of ferrozine solution, was used as a sample blank, which is required for error correction (unequal color of the sample solutions). Methanol, instead of a sample solution, was also used as a control. Results were expressed as relative iron chelating activity compared to the unchelated (without ferrozine) Fe<sup>2+</sup> reaction, and EDTA was used as the reference standard. The FIC activity was calculated using the following equation: FIC activity (%) = (A<sub>0</sub> – (A<sub>1</sub> – A<sub>2</sub>))/A<sub>0</sub> × 100, where A<sub>0</sub> was the Abs of the control, A<sub>1</sub> was the Abs of the sample or standard, and A<sub>2</sub> was the Abs of the blank.

### 2.5. Determination of the Total Phenolic and Total Flavonoid Contents

Total phenolic content (TPC) was determined by using the Folin–Ciocalteu colorimetric methodology according to the method of Wang et al. [24] with some modifications [21]. An aliquot of 100 µL of each extract sample (2 mg/mL) plus 1500 µL of distilled water and 100 µL of 2 N FCR (Folin–Ciocalteu reagent) was homogenized in a vortex for 15 s and placed in the dark for 3 min. After that, 300 µL of Na<sub>2</sub>CO<sub>3</sub> (10% w/v) was homogenized and incubated for 5 min at 50 °C. The absorbance (Abs) of the samples was measured at 760 nm. Gallic acid was used to produce a standard calibration curve at various concentrations, and the results were expressed in milligrams of gallic acid equivalents per gram of dried extract (mg GAE/g of DE).

The total flavonoid content was determined by the aluminum chloride colorimetric method [25] with some modifications [21]. An aliquot of 100 µL of each extract sample (2 mg/mL) was mixed with 100 µL of AlCl<sub>3</sub> (10%, w/v), 100 µL of KCH<sub>3</sub>CO<sub>2</sub> (10% w/v), and 900 µL of distilled water. The absorbance was measured at 415 nm after 30 min at room temperature. Rutin was used to produce a standard calibration curve at various concentrations, and the results were expressed as mg of rutin equivalents per gram of dried extract (mg RE/g of DE).

### 2.6. Statistical Analysis

All determinations were expressed as mean ± standard deviation (SD) and performed in triplicate. The one-way analysis of variance test (ANOVA) was carried out to assess and indicate any significant differences between the mean values obtained from each sample. Correlations between the evaluated parameters were obtained using Pearson’s correlation coefficient (r). Significance was based on a confidence level of 95% (p < 0.05). The statistical analysis was performed using SPSS 20.0 (SPSS Inc., Chicago, IL, USA).

## 3. Results and Discussion

### 3.1. Yield Determinations of Aromatic Medicinal Plants and *C. sinensis* Green Tea Extracts

The yield percentages are described in Table 2. The highest yield percentage was observed for *Hibiscus rosa-sinensis* (60.90%), followed by *Stevia rebaudiana* (45.56%), *Camellia sinensis* (39.50%), and *Mentha pulegium* (34.80%). The lowest yield percentage was observed for *Pimpinella anisum* (16.53%).

**Table 2.** Antioxidant activities (free radical scavenging activity—FRSA, ferric reducing antioxidant power—FRAP, and ferrous ion chelating—FIC) in aromatic medicinal plants and Azorean *Camellia sinensis* (green tea) \*.

Aromatic Medicinal Plants	Yield (%)	FRSA (EC <sub>50</sub> , µg/mL)	FRAP (EC <sub>50</sub> , µg/mL)	FIC (%)
<i>Aloysia citrodora</i>	30.90	14.14 ± 0.14 <sup>b</sup>	11.42 ± 0.18 <sup>c</sup>	51.90 ± 0.88 <sup>e</sup>
<i>Cymbopogon citratus</i>	19.87	35.30 ± 0.61 <sup>d</sup>	39.50 ± 0.05 <sup>f</sup>	80.60 ± 1.43 <sup>b</sup>
<i>Mentha pulegium</i>	34.80	14.22 ± 0.23 <sup>b</sup>	12.07 ± 0.14 <sup>c</sup>	53.71 ± 0.42 <sup>e</sup>

Table 2. Cont.

Aromatic Medicinal Plants	Yield (%)	FRSA (EC <sub>50</sub> , µg/mL)	FRAP (EC <sub>50</sub> , µg/mL)	FIC (%)
<i>Pimpinella anisum</i>	16.53	94.48 ± 0.17 <sup>e</sup>	75.00 ± 0.14 <sup>h</sup>	36.71 ± 2.77 <sup>g</sup>
<i>Stevia rebaudiana</i>	45.56	20.40 ± 0.91 <sup>c</sup>	15.10 ± 0.14 <sup>c,d</sup>	42.80 ± 2.51 <sup>f</sup>
<i>Lavandula angustifolia</i>	27.57	23.43 ± 0.25 <sup>c</sup>	11.11 ± 0.06 <sup>c</sup>	58.11 ± 1.59 <sup>d</sup>
<i>Thymus citriodorus</i>	24.49	24.10 ± 0.01 <sup>c</sup>	12.19 ± 0.13 <sup>c</sup>	68.48 ± 0.24 <sup>c</sup>
<i>Achyrocline satureioides</i>	26.49	16.27 ± 0.27 <sup>b</sup>	7.85 ± 0.07 <sup>b</sup>	51.86 ± 1.11 <sup>e</sup>
<i>Salvia rosmarinus</i>	19.44	15.48 ± 0.03 <sup>b</sup>	8.13 ± 0.17 <sup>b</sup>	69.44 ± 0.66 <sup>c</sup>
<i>Laurus nobilis</i>	25.24	22.89 ± 0.17 <sup>c</sup>	28.93 ± 0.45 <sup>e</sup>	77.76 ± 1.15 <sup>b</sup>
<i>Hibiscus rosa-sinensis</i>	60.90	92.55 ± 0.35 <sup>e</sup>	55.76 ± 0.69 <sup>g</sup>	4.65 ± 0.43 <sup>h</sup>
<i>Camellia sinensis</i> (green tea)	39.50	3.43 ± 0.06 <sup>a</sup>	5.12 ± 0.21 <sup>a</sup>	61.10 ± 0.34 <sup>d</sup>
Ascorbic acid	-	2.11 ± 0.04 <sup>a</sup>	3.91 ± 0.27 <sup>a</sup>	-
EDTA	-	-	-	98.16 ± 0.15 <sup>a</sup>

\* Values are mean ± SD (n = 3). Different superscript letters indicate that values are significantly different (p < 0.05). Half-maximal effective concentration—EC<sub>50</sub>. Ethylenediamine–tetra acetic disodium salt—EDTA.

### 3.2. Antioxidant Activity Determination

Several methods were used to study the antioxidant properties of the medicinal plant extracts. Antioxidant activity should not be investigated based on a single antioxidant assay for more complete/accurate results. To estimate the antioxidant properties of bioactive natural compounds, the samples under study were evaluated using different in vitro antioxidant assays, such as DPPH-FRSA, FRAP, and FIC activity (Table 2).

Based on the DPPH Free Radical Scavenging Activity (FRSA), the results revealed that *Camellia sinensis* showed a higher ability to scavenge DPPH radicals than the other aromatic medicinal plants, with an EC<sub>50</sub> of 3.43 µg/mL, followed by *Aloysia citrodora* and *Mentha pulegium*, *Salvia rosmarinus*, *Achyrocline satureioides*, *Stevia rebaudiana*, *Laurus nobilis*, *Lavandula angustifolia*, and *Thymus citriodorus*, with values of 14.14 and 14.22, 15.48, 16.27, 20.40, 22.89, 23.43, and 24.10 µg/mL, respectively. Furthermore, *Hibiscus rosa-sinensis* and *Pimpinella anisum* presented lower abilities to scavenge DPPH radicals, with EC<sub>50</sub> values of 92.55 and 94.48 µg/mL, respectively. According to Rita et al. [15], *Aloysia citrodora* presented an EC<sub>50</sub> of 390 µg/mL, and Yakoubi et al. [26] showed an EC<sub>50</sub> of 12.84 µg/mL for *Mentha pulegium*. Brindisi et al. [27] obtained a value between 8.80 and 14.76 µg/mL for *Salvia rosmarinus*. Sharman and Thakur [28] observed an EC<sub>50</sub> value of 117.23 µg/mL for *Hibiscus rosa-sinensis*, and Farzaneh et al. [29] obtained a value of 98.47 µg/mL for *Pimpinella anisum*; these values are all similar to our results. However, other authors have presented different EC<sub>50</sub> for *Achyrocline satureioides* (112.6 µg/mL) [30], *Laurus nobilis* (90–130 µg/mL) [31], and *Thymus citriodorus* (69.39 µg/mL) [32]. Moreover, all the samples presented in this study showed lower antioxidant capacities than ascorbic acid (EC<sub>50</sub> = 2.11 µg/mL).

The FRAP of each extract was evaluated based on their abilities to reduce ferric iron (Fe<sup>3+</sup> complex to Fe<sup>2+</sup>). For FRAP results of aromatic medicinal plants and *Camellia sinensis* green tea samples, expressed as EC<sub>50</sub> values (µg/mL), this study shows that *Camellia sinensis* presented the best EC<sub>50</sub> value (5.12 µg/mL), followed by *Achyrocline satureioides* (7.85 µg/mL) and *Salvia rosmarinus* (8.13 µg/mL), and significantly lower FRAP values (higher EC<sub>50</sub>) were observed for *Pimpinella anisum*, *Hibiscus rosa-sinensis*, *Cymbopogon citratus*, and *Laurus nobilis* (75, 55.76, 39.50, and 28.93 µg/mL, respectively). However, some aromatic plants also presented interesting FRAP values, as *Lavandula angustifolia*, *Aloysia citrodora*, *Mentha pulegium*, *Thymus citriodorus*, and *Stevia rebaudiana* had EC<sub>50</sub> values between 11.11 and 15.10 µg/mL. Similar results for *Mentha pulegium* (12.25 µg/mL) were reported by Yakoubi et al. [26] compared to our results.

Conversely, some authors presented lower FRAP measurements for *Achyrocline satureioides* (84.8 µg/mL) [30], *Cymbopogon citratus* (210 µg/mL) [33], *Laurus nobilis* (90–120 µg/mL) [31], *Aloysia citrodora* (205 µg/mL), and *Thymus citriodorus* (228 µg/mL) [15] as compared to our results.

Regarding the FIC, the chelating ability of each extract at various concentrations was determined by measuring the inhibition of the  $\text{Fe}^{2+}$ –ferrozine complex formation. For FIC activity, *Cymbopogon citratus*, *Laurus nobilis*, *Salvia rosmarinus*, and *Thymus citriodorus* exhibited the highest values (80.60, 77.76, 69.44, and 68.48%, respectively), while *Hibiscus rosa-sinensis* exhibited the lowest value (4.65%). *Camellia sinensis* presented a value of 61.10%. However, the higher values were lower than that of EDTA (98.16%), a potent metal-ion chelator.

### 3.3. Total Phenolic and Total Flavonoid Contents (TPC and TFC)

Table 3 presents the TPC and TFC values found for the aromatic medicinal plants and *Camellia sinensis* green tea samples. TPC and TFC are known as polyphenolic compounds and are also recognized as antioxidants, and their consumption is increasing every day because of their beneficial effects on health [34].

**Table 3.** Total phenolic and total flavonoid contents in dry extracts (DEs) of aromatic medicinal plants and Azorean *Camellia sinensis* (green tea) \*.

Aromatic Medicinal Plants	TPC (mg GAE/g DE)	TFC (mg RE/g DE)
<i>Aloysia citrodora</i>	187.15 ± 1.11 <sup>b</sup>	204.64 ± 1.47 <sup>b</sup>
<i>Cymbopogon citratus</i>	82.85 ± 2.19 <sup>e</sup>	49.74 ± 1.46 <sup>g</sup>
<i>Mentha pulegium</i>	199.15 ± 1.23 <sup>b</sup>	92.84 ± 1.11 <sup>e</sup>
<i>Pimpinella anisum</i>	47.37 ± 1.15 <sup>f</sup>	28.08 ± 1.03
<i>Stevia rebaudiana</i>	163.66 ± 2.55 <sup>c</sup>	74.55 ± 1.42 <sup>f</sup>
<i>Lavandula angustifolia</i>	99.09 ± 0.75 <sup>e</sup>	16.45 ± 1.47 <sup>i</sup>
<i>Thymus citriodorus</i>	157.04 ± 2.25 <sup>c,d</sup>	168.39 ± 1.27 <sup>c</sup>
<i>Achyrocline satureioides</i>	179.81 ± 1.09 <sup>b,c</sup>	265.75 ± 1.10 <sup>a</sup>
<i>Salvia rosmarinus</i>	186.31 ± 0.92 <sup>b</sup>	132.84 ± 1.10 <sup>d</sup>
<i>Laurus nobilis</i>	147.98 ± 0.35 <sup>d</sup>	30.89 ± 2.13 <sup>h</sup>
<i>Hibiscus rosa-sinensis</i>	28.98 ± 0.19 <sup>g</sup>	20.20 ± 0.64 <sup>i</sup>
<i>Camellia sinensis</i> (Green tea)	294.43 ± 2.5 <sup>a</sup>	49.90 ± 1.72 <sup>g</sup>

\* Values are mean ± SD ( $n = 3$ ). Different superscript letters indicate that values are significantly different ( $p < 0.05$ ). Gallic acid equivalents—GAE. Rutin equivalents—RE.

The TPC results, expressed in milligrams of GAE/g DE, were higher in *Camellia sinensis* green tea, followed by *Mentha pulegium*, *Aloysia citrodora*, *Salvia rosmarinus*, *Achyrocline satureioides*, *Stevia rebaudiana*, *Thymus citriodorus*, and *Laurus nobilis*, with values of 294.43, 199.15, 187.15, 186.31, 179.81, 163.66, 157.04, and 147.98 mg GAE/g DE, respectively. However, lower values were obtained for *Lavandula angustifolia*, *Cymbopogon citratus*, *Pimpinella anisum*, and *Hibiscus rosa-sinensis*, recorded as 99.09, 82.85, 47.37, and 28.98 mg GAE/g DE. Our results presented higher values than those of some studies observed from other authors [8,31,35,36]. Moreover, Sharma and Thakur [28] revealed a higher value for *Hibiscus rosa-sinensis* (54.75 mg GAE/g DE) as compared to our study.

The TFC results, expressed in milligrams of RE/g DE, were higher in *Achyrocline satureioides*, followed by *Aloysia citrodora*, *Thymus citriodorus*, *Salvia rosmarinus*, *Mentha pulegium*, and *Stevia rebaudiana*, with values of 265.75, 204.64, 168.39, 132.84, 92.84, and 74.55 mg RE/g DE. Conversely, *Hibiscus rosa-sinensis* and *Lavandula angustifolia* presented the lowest values of 20.20 and 16.45 mg RE/g DE, respectively. As compared to our results, some authors presented lower values for *Thymus vulgaris* (44.20 mg QE/g DE) [37] and *Lavandula angustifolia* (3.37–4.85 mg QE/g DE) [38]. However, Yakoubi et al. [26] showed a higher value for *Mentha pulegium* (155.83 mg QE/g DE), and Mak et al. [39] observed a similar result for *Hibiscus rosa-sinensis*, but a lower value was detected in a study by Sharma and Thakur [28], with values of 27.68 and 14.11 mg QE/g DE, respectively, as compared to our results.

### 3.4. Pearson Correlation between Parameters

A significant correlation was observed only among two methods used to determine the biological activities (Table 4). A very strong positive correlation was observed between FRSA and TPC ( $r = 0.937$ ); however, a weak correlation was found for FRSA and FIC activity ( $r = 0.356$ ), for TPC and FIC activity ( $r = 0.428$ ), and for TPC and TFC ( $r = 0.414$ ).

**Table 4.** Correlation matrix of the studied parameters of aromatic medicinal plants and Azorean *Camellia sinensis* (green tea) (Pearson correlation coefficients).

	FRSA	FIC Activity	TPC	TFC
FRSA	1	–	–	–
FIC activity	0.356	1	–	–
TPC	0.937	0.428	1	–
TFC	0.201	0.150	0.414	1

Free radical scavenging activity—FRSA; ferrous ion chelating—FIC; total phenolic content—TPC; total flavonoid content—TFC.

These results emphasize that polyphenols were the compounds that contributed most to the antioxidant activities in all studied samples. Regarding the FIC activity, the correlations observed in the present study may suggest that compounds other than polyphenols are also capable of binding Fe(II) ions and may also contribute to the observed correlation.

## 4. Conclusions

In the present study, eleven aromatic medicinal plants and *Camellia sinensis*, cultivated in Azores Islands, were investigated for their phytochemical and biological activities, including antioxidant capacities and total phenolic and flavonoid contents. The data in this study revealed differences in the biological activities of aromatic medicinal plants as compared to *Camellia sinensis* green tea that could be due to the effects of harvesting time, location, moderate climate conditions, and genetic factors of each aromatic medicinal plant.

The results revealed that *Camellia sinensis* showed higher values for free radical scavenging activity (FRSA), ferric reducing antioxidant power (FRAP), and total phenolic content (TPC). However, some aromatic medicinal plants also presented significant results in terms of FRSA and FRAP, particularly *Aloysia citrodora*, *Mentha pulegium*, and *Stevia rebaudiana*. *Cymbopogon citratus* and *Laurus nobilis* presented higher levels of ferrous ion-chelating (FIC) activity. For total flavonoid content (TFC), *Achyrocline satureioides* showed the highest value. The strong correlations between antioxidant activity, determined by DPPH assays, and the total phenolic content confirm that phenolic compounds contribute significantly to the antioxidant properties of aromatic medicinal plants. Due to the potential antioxidant activity, some of the aromatic medicinal plants and their derivatives can be used to replace synthetic food preservatives and/or antioxidants that have negative effects on human health. These findings support the view that certain aromatic medicinal plants are potential candidates for use in the cosmetic, pharmaceutical, and food industries, mainly for their content of phenolics and flavonoids.

The combination of aromatic medicinal plants with Azorean *Camellia sinensis* can be beneficial in terms of providing extra flavor and, in some cases, enhancing the value of some components, such as flavonoid compounds. However, based on these findings, the present study is an attempt to update our knowledge about the different therapeutic applications of several medicinal plant products for various pharmacological targets, and further analysis should be performed to obtain more understandable information about aromatic medicinal plants and their potential uses for human health.

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**Data Availability Statement:** The data that support the findings of this study are available upon request from the corresponding author.

**Conflicts of Interest:** Author Madalena Hintze Motta was employed by the company Gorreana Tea Plantation. The remaining 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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