horticulturae

Article

Evaluation of the Quality and Antioxidant Activity of Dehydrated Medicinal Herbs

Adna Prado Massarioli [1], Severino Matias de Alencar [1], Adriano Francisco Siqueira [2], Mariana Pereira de Melo [2], Igor Gomes Vidigal [2] and Ana Lúcia Gabas Ferreira [2,*]

[1] Department of Agri-Food Industry, Food and Nutrition, "Luiz de Queiroz" College of Agriculture, University of Sao Paulo, Avenida Pádua Dias, Piracicaba 13418-900, SP, Brazil; adnaprado@usp.br (A.P.M.); smalencar@usp.br (S.M.A.)

[2] Department of Basic and Environmental Sciences, Engineering School of Lorena, University of Sao Paulo, Estrada Municipal do Campinho No. 100, Lorena 12602-810, SP, Brazil; adrianoeel@usp.br (A.F.S.); marianapm@usp.br (M.P.M.); igor.vidigal@alumni.usp.br (I.G.V.)

* Correspondence: gabas@usp.br

Abstract: This study aims to evaluate the effects of drying in a forced-air oven or solar dryer on the drying rates, physicochemical and microbiological characteristics, and antioxidant properties of rosemary, mint, common fennel, lemon grass, and basil. The drying rates of all herbs were higher in the forced-air oven in comparison to the solar dryer. According to results obtained for herbal properties after this different drying process, mint was less affected by both drying conditions. On the other hand, regardless of the method of drying used, all dried herbs exhibited similar antioxidant properties, mainly due to the presence of total phenolics. The antioxidant activities of oven-dried herbs ranged from 19.18 to 71.55% and increased in the order common fennel < lemon grass < mint < basil < rosemary, while the activities of sun-dried samples varied from 17.73 to 58.27% and increased in the order basil < common fennel < lemon grass < mint < rosemary. The results obtained demonstrate that the process of drying can alter the quality of an herbal product, implying that standardization of post-harvest steps is essential to ensure the consistency of an herbal product.

Keywords: antioxidant activity; drying processes; medicinal herbs; phenolic compounds

Citation: Massarioli, A.P.; Alencar, S.M.; Siqueira, A.F.; Melo, M.P.; Vidigal, I.G.; Ferreira, A.L.G. Evaluation of the Quality and Antioxidant Activity of Dehydrated Medicinal Herbs. *Horticulturae* **2023**, *9*, 597. https://doi.org/10.3390/horticulturae9050597

Academic Editor: Bastien Christ

Received: 18 March 2023
Revised: 4 May 2023
Accepted: 15 May 2023
Published: 18 May 2023

1. Introduction

Over the past three decades, there has been a constant, and at times, exponential growth in global interest in the use of herbal medicines. This increase in the popularity and usage of herbal medicines is evident in the global market. The global herbal medicine market size was valued at USD 151.91 billion in 2021, which is mainly concentrated in Asia, North America, and Europe [1]. In Brazil, however, research and development of herbal remedies has suffered from insufficient funding, and the trade represents less than 5% of the total Brazilian medicinal market. Thus, despite the large number of therapeutic agents that originate exclusively from the immensely diverse Brazilian flora, many of the valuable active principles have been patented and developed by foreign companies or government agencies.

The development time and production costs of herbal medicines can be, in some cases, less than those of semi-synthetic or synthetic agents. Hence, herbal products are typically more accessible to low-income populations by virtue of their lower price, and their use is often rooted in local culture and tradition. Moreover, since herbal medicines are considered natural products, they are not subject to the regulations imposed on prescription drugs and are readily available in drugstores, health food shops, and supermarkets, and can be grown by those who have the minimum conditions required. Because of such liberal policies, self-medication with products prepared and dispensed by herbalists has become popular all over the world, thereby reducing the demand for assistance from health professionals in

the treatment of common ailments. However, the increasing growth in the use of herbal medicines has raised concerns about their efficacy and safety, particularly regarding the assessment of therapeutic doses and the quality and toxicity of the products [2].

In recognition of the insufficiency in quality control of herbal medicines, the World Health Organization (WHO) has developed technical guidelines relating to the cultivation and collection of medicinal plants and the recording and documentation of data during processing [3]. The WHO encourages countries to carry out regional surveys of the species used in traditional folk medicine to advise against those that may be harmful and to select those that have proven efficacy. Moreover, since appropriate agricultural and collection practices, along with post-harvest processing of selected medicinal plants, are fundamental for the safety and efficacy of herbal products [4], the WHO has stimulated the development of cultivation projects. The Brazilian Ministry of Health authorized the use of herbal medicines in the national health care system (Sistema Único de Saude—SUS) in 1988 and issued appropriate guidelines in 2001, despite the practice not being recognized by the National Medical Council of Brazil. The Brazilian government also supports projects relating to the production of medicinal plants by family farms and rural settlements as a strategy to generate income, reduce regional inequalities, and scale down the cost of health care [5,6].

Plant species may present sound therapeutic properties even if their specific active principles have not been fully elucidated. For example, various herbs and spices are targeted by the food industry because they are important for food preservation as well as being beneficial to health, and such properties can be attributed to the antioxidant and free-radical scavenging activities of the phenolic constituents. Many compounds of this class are produced by plants as defense mechanisms, and their antioxidant activities impede microbial invasion, a property that can help to prevent the putrefaction of food products and, thereby, extend their shelf life. Phenolics are also key constituents of many functional foods since they exert a protective effect against inflammatory and degenerative diseases [7].

The preservation of active principles of herbs and spices depends on the care given to the plant material at all stages of production, from cultivation through post-harvest processing. Since most medicinal plants are marketed in their dehydrated form, the procedure employed in drying the fresh material is critical to the quality of the final product. While the literature on the separation, purification, and identification of antioxidants is vast [8–12], little has been published concerning the quality (organoleptic characteristics and therapeutic effects) of medicinal plants following post-harvesting processing.

Considering the above, we hypothesized that the use of a solar dryer rather than a forced-air oven would affect the final quality of an herbal product. In order to test this hypothesis, five herbs used as remedies and food condiments were subjected to the two drying processes, and the drying rates, physicochemical and microbiological characteristics, and antioxidant properties were analyzed. The selection criteria of plants in this study were herbs available, cultivated, produced, and consumed in the region.

2. Materials and Methods

2.1. Origin of Plant Material

Fresh leaves of mint (*Mentha x piperita* L.; Lamiaceae), common fennel (*Foeniculum vulgare* Mill.; Apiaceae), lemon grass [*Cymbopogon citratus* (DC) Stapf.; Poaceae], rosemary (*Rosmarinus officinalis* L.; Lamiaceae), and basil (*Ocimum basilicum* L.; Lamiaceae) were collected from the medicinal garden of Sao Paulo University. Voucher specimens were verified at the Herbarium ESA at the Department of Biological Sciences, Luiz de Queiroz College of Agriculture of the University of São Paulo: *Ocimum basilicum* (voucher 051210); *Mentha piperita* (voucher 016650); *Foeniculum vulgare* convar. *azoricum* (voucher 083960); *Cymbopogon citrattus* (voucher 004981); and *Rosmarinus officinalis* (voucher 050997).

2.2. Forced-Air Oven Drying

Fresh leaves of each of the five species were placed in pre-weighed trays (n = 3 per herb), and leaf masses were determined using a UX420H semi-analytical scale (Shimadzu, Kyoto, Japan) with an accuracy of 0.001 g. The trays were transferred to a TE-394/4-MP forced-air oven (Tecnal, Piracicaba, Brazil) maintained at 50 °C with an airflow rate estimated at ca. 1.0 m/s and drying kinetics were established by determining the mass of leaves in one of the trays at 15 min intervals until a constant dry weight was attained. The drying temperature was chosen because it was not high enough to cause degradation of the active principles but was similar to the maximum average air temperature achieved by the solar dryer so that the two drying processes could be performed under equivalent conditions for the purpose of comparison.

2.3. Solar Drying

The solar dryer (Figure 1) featured the following components: (A) an air heating compartment comprising a rectangular duct with a glass top and absorber plate to collect and store solar radiation, where the duct and absorber plate were formed from 1 mm thick aluminum sheets, and the side walls and base were isolated with Styrofoam; (B) a drying compartment (1.5 m high × 0.5 m × 0.5 m) fabricated from carbon steel and wood and equipped with drying trays; and (C) a turbine roof ventilator. Air was heated by sunlight as it passed between the glass panel and the absorber plate in the heating compartment and was drawn through the drying compartment by the turbine action of the roof ventilator. Fresh leaves of each of the five herbs were placed in pre-weighed trays (n = 2 per herb) and transferred to the solar dryer, which was exposed to solar radiation all day long. The mass of leaves in one of the trays was determined using semi-analytical scales at 45 min intervals until a constant dry weight was attained, and air temperature and relative humidity were recorded at each of the weighing procedures by using a digital thermo-hygrometer (Incoterm) installed inside the solar dryer. Experiments involving different herbs were performed on different days, and at night, the dryer was moved into the unit operations laboratory.

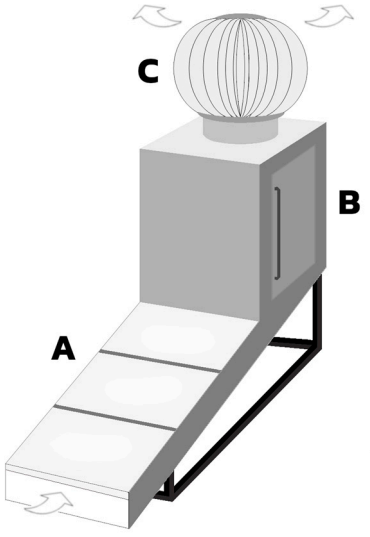

Figure 1. Solar dryer showing (**A**) air heating compartment, (**B**) drying compartment, and (**C**) turbine roof ventilator.

2.4. Drying Curves

The moisture ratio as a function of drying time was calculated according to Equation (1), while the goodness of fit of the experimental data was tested using Page's mathematical model shown in Equation (2) [13]:

$$M = \frac{X - X_{eq}}{X_0 - X_{eq}} \quad (1)$$

$$M = C\exp(-kt^n) \quad (2)$$

where M is the ratio dimensionless moisture; X is the moisture content at a certain time t; X_0 and X_{eq} are the initial and equilibrium moisture contents, respectively; and C, k, and n are the Page drying coefficients.

The drying rate [(kg water removed/kg dry matter)/min] was calculated according to Equation (3):

$$\text{Drying rate} = \frac{M_{t+dt} - M_t}{dt} \quad (3)$$

where M_t is the moisture content at time t, $M_{t\,+\,dt}$ is the moisture content at time $t + dt$, and t is the drying time interval.

2.5. Physicochemical and Microbiological Methods

Dried herbs were characterized according to the content of dry matter, minerals, protein, fiber, reducing sugars, and sucrose using methods described by the Association of Official Analytical Chemists [14]. All assays were performed in duplicate.

The identification and counting of micro-organisms followed standard methods: (i) molds and yeast—solid agar surface plating method [15]; (ii) *Staphylococcus aureus*—direct plating technique [16]; (iii) sulfite-reducing clostridia—tryptose sulfite cycloserine (TSC) agar method [17]; (iv) *Salmonella* sp.—non-selective pre-enrichment followed by selective enrichment in tetrathionate and selenite cystine broths [18]; and (v) total and fecal coliforms—most probable number (MPN) method in multiple fermentation tubes [19].

The colors of dried herb samples were assessed using a MiniScan XE Plus colorimeter (HunterLab, Reston, VA, USA) and reported according to the Commission Internationale de l'Eclairage L*a*b* color space method.

2.6. Phytochemical Methods

Extracts of leaves from each of the herbs were obtained by resuspending 10 g of pulverized material in 100 mL methanol and incubating at 50 °C in a water bath for 15 min. Suspensions were sieved through qualitative filter paper, and the filtrate was concentrated in a Tecnal (Piracicaba, SP, Brazil) model TE-211 rotary evaporator. Extracts of herbs were stored at −20 °C until required for analysis, and all assays were conducted in duplicate.

Total phenolics in the extracts were quantified according to the Folin–Ciocalteu method [20]. The herb extract was dissolved in methanol and incubated with the reagent for 120 min at room temperature, following which absorbance at 740 nm was recorded on a Shimadzu (Kyoto, Japan) UV-mini 1240 spectrophotometer. The phenolics content was calculated as gallic acid content (mg/g) from a calibration curve.

The concentration of flavonoids was determined using the method described by Park et al. [21], with minor modifications. A total of 0.5 mL of herb extract, 4.3 mL of 80% ethanol, 0.1 mL of 10% Al(NO3)3, and 0.1 mL of 1M potassium acetate solutions were added. The mixture reacted for 40 min, and the absorbance of the mixture solution was measured at 415 nm. Total flavonoid content was calculated as quercetin (mg/g) from a calibration curve.

The antioxidant activity of a concentrated extract dissolved in methanol was determined spectrophotometrically by monitoring the coupled autoxidation of β-carotene and linoleic acid in the presence of analyte at 470 nm every 20 min over a 120 min period as

described by Emmons et al. [22]. Butylated hydroxytoluene and α-tocopherol were used at concentrations of 90 µg/mL as positive controls, while methanol was employed as a negative control. Antioxidant activity was expressed as percentage inhibition relative to the control after 120 min according to Equation (4):

$$\text{Antioxidant activity} = \frac{(DR_c - DR_s)}{DR_c} \times 100 \quad (4)$$

where the degradation rate of the negative control (DR_c) and that of the extract or positive control (DRs) were calculated from:

$$\text{Degradation rate} = {}^{\ln(\text{absorbance at 0 min}/\text{absorbance at 120 min})}\Big/_{120} \quad (5)$$

All assays were conducted in duplicate.

2.7. Statistical Analysis

Data relating to physicochemical and phytochemical characteristics were analyzed using analysis of variance (ANOVA) followed by Tukey's test. All analyzes were performed using Statistica™ version 13.0 (TIBCO Software Inc, Palo Alto, CA, USA) software.

3. Results and Discussion

3.1. Drying Curves

Moisture ratios as a function of drying time for the herbs rosemary, lemon grass, common fennel, mint, and basil are shown in Figure 2 (note that the *x*-axis has different limits; the drying time varies in Figure 2a) from 0 to 400 min and in Figure 2b) from 0 to 3600 min). The reduction in the moisture ratio was considerably faster when plants were dried in a forced-air oven (Figure 2a) rather than in a solar dryer (Figure 2b), with oven drying completed in 300 min while solar drying took 1800 min or more. This divergence can be explained by the different drying conditions applied since a constant temperature of 50 °C was maintained inside the oven while that in the solar dryer fluctuated but never exceeded 45 °C. The maximum–minimum values of temperature and relative humidity, respectively, inside the solar dryer for the studied herbs were 45–26 °C and 46–35%. The moisture ratio of the rosemary herb was much higher than those of the others at any given time.

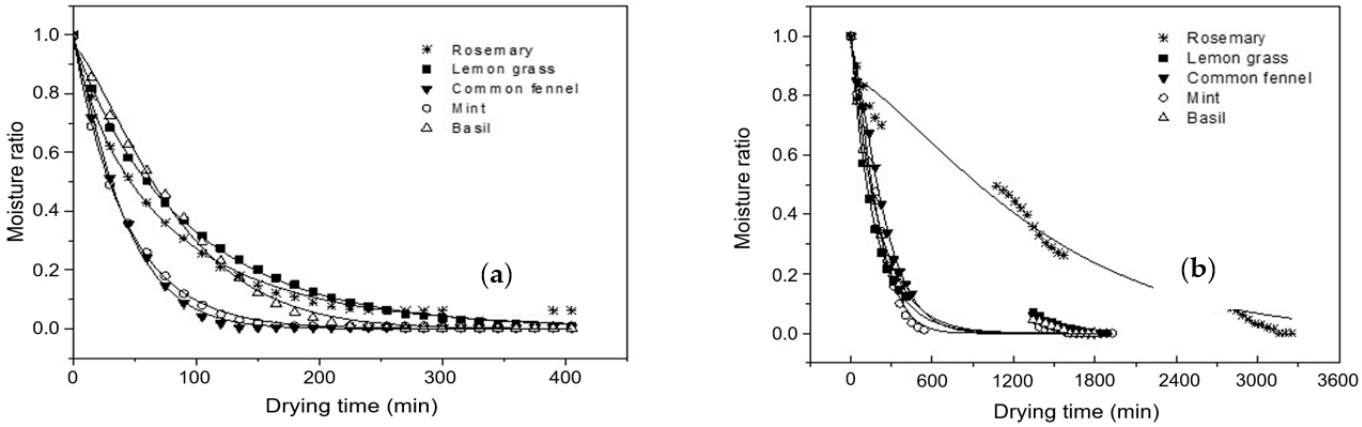

Figure 2. Variation in moisture ratios of herbs as a function of drying time in (**a**) a forced-air oven at 50 °C and (**b**) a solar dryer at 26 °C min–45 °C max.

The data presented in Table 1 confirm that Page's model (Equation (2)) satisfactorily described the reductions in moisture ratio as a function of time for the two drying processes with high coefficients of determination (R^2) in the range 0.972 to 0.998 for the various herbs tested. The R^2 value for Page's best-fit curve obtained for mint dried in a forced-air oven at

50 °C with an air velocity of 1.0 m/s was similar to that reported (0.9987) by Doymaz [23] for mint leaves dried in a laboratory cabinet dryer at 55 °C. However, according to this author, the highest R^2 value (0.9996) for a dryer operated at 55 °C could be obtained using a logarithmic model. Nevertheless, Park et al. [24] applied Page's model to describe the moisture ratio vs. time curve for mint dried in a forced-air oven at 50 °C with an air velocity of 1.0 m/s and reported model constants of k (10^2) = 3.70, n = 0.98 and R^2 = 1.0, values that were not dissimilar to those reported in Table 1. In addition, Akpinar [25] employed Page's model to describe the time-related reductions in moisture ratio for various herbs dried in direct sunlight and obtained model constants of k (10^2) = 0.029, n = 0.934 and R^2 = 0.985 for mint and k (10^2) = 0.041, n = 0.896 and R^2 = 0.995 for basil. Comparing our results (Table 1) with those reported by Akpinar [25], the n values for both herbs and the k values of mint were essentially similar, while the k values for basil differed by an order of magnitude.

Table 1. Goodness of fit of the experimental data described in terms of Page's mathematical model.

Drying Process	Page's Model Parameters	Herbs				
		Rosemary	Lemon Grass	Common Fennel	Mint	Basil
Oven-Dried	C	0.988	0.988	0.988	0.993	0.966
	k (10^2)	1.49	1.30	1.09	2.03	0.26
	n	1.01	0.97	1.20	1.04	1.32
	R^2	0.997	0.988	0.994	0.998	0.996
Sun-dried	C	0.862	1.01	0.976	0.965	0.990
	k (10^2)	0.006	0.82	0.057	0.063	0.449
	n	1.33	0.932	1.34	1.38	1.01
	R^2	0.972	0.991	0.994	0.995	0.993

Plots of drying rate vs. moisture ratio (Figure 3a) revealed that the highest rates were typically attained during the oven drying of common fennel [maximum rate 0.0065 (kg water/kg dry matter)/min] while the time required to attain equilibrium moisture content for this herb was 150 min. In contrast, lemon grass presented the lowest drying rates [maximum rate 0.0030 (kg water/kg dry matter)/min], and the equilibrium moisture content was reached after approximately 320 min. Interestingly, the moisture content of rosemary in natura was higher in comparison with that of common fennel, and the maximum drying rate was higher [0.007 (kg water/kg dry matter)/min]. However, the drying rate of rosemary exhibited large variations, and at moisture ratios lower than 0.6 (dry basis), the drying rate was comparable with that of mint [0.0035 (kg water/kg dry matter)/min]. Doymaz [23] reported that mint dried in a cabinet dryer at 45 °C exhibited a drying rate of 0.03 (kg water/kg dry matter)/min at a moisture ratio of 0.6 (dry basis), although the 10-fold higher drying rate recorded by this author could be attributed to the higher air velocity (4.1 m/s) employed.

Under the conditions of the solar dryer, the drying rates of lemon grass surpassed those of all other herbs when moisture ratios were higher than ~0.65 (dry basis) and attained a maximum value of approximately 0.0032 (kg water/kg dry matter)/min, although mint exhibited the highest drying rates at lower moisture ratios (Figure 3b). The lowest drying rate was observed for rosemary, a feature that may be attributed to the weather conditions on the day of the experiment.

Comparing the two processes, the drying rates of all herbs were much higher in the forced-air oven than in the solar dryer. It is interesting to note, however, that lemon grass exhibited the slowest drying rate in the oven but the highest in the solar dryer.

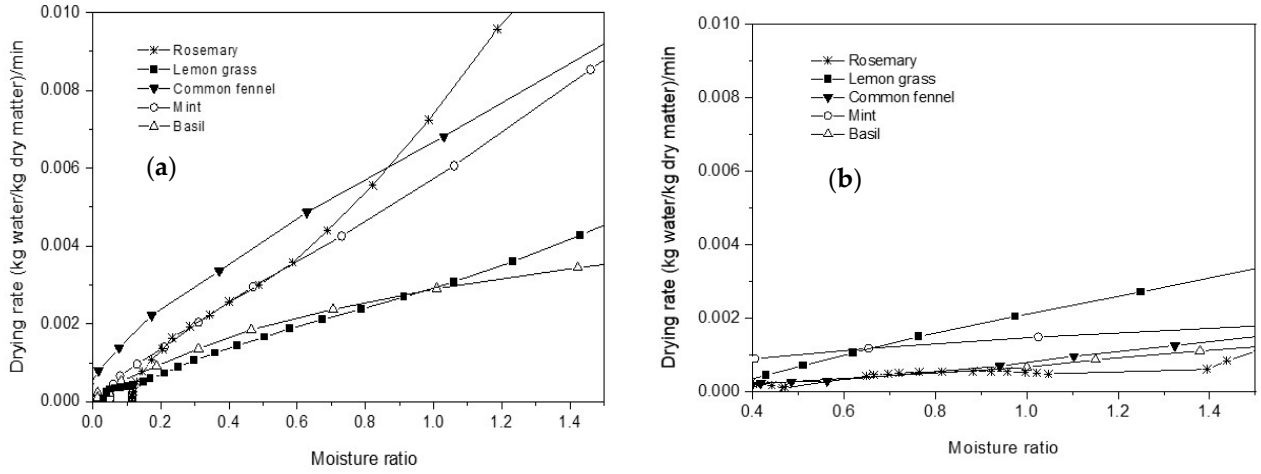

Figure 3. Variation in drying rate of herbs as a function of moisture ratio during drying in (**a**) a forced-air oven at 50 °C and (**b**) a solar dryer at 26 °C min–45 °C max.

3.2. Physicochemical Characteristics of Oven- and Sun-Dried Herbs

Table 2 shows the centesimal composition of the oven- and sun-dried herbs in the dry basis and the moisture content in the wet basis. For most of the herbs, the method of drying affected the centesimal composition significantly ($p < 0.05$) but to different extents. Thus, all the physicochemical parameters of basil were affected significantly by the drying process, whereas in the case of mint, all parameters, except reducing sugar content, were similar regardless of the method of drying. The most plausible explanation for the variations observed in basil is that the herb samples could have been affected by environmental factors [26].

Table 2. Centesimal composition of oven- and sun-dried herbs in dry basis and moisture in wet basis.

Herbs	Drying Process	Composition [1] (g/100 g)					
		Moisture	Mineral Content	Protein	Fiber	Reducing Sugars	Sucrose
Rosemary	Oven-dried	88.81 ^{a}	7.01 ^{a}	10.90 ^{a}	21.32 ^{a}	6.58 ^{a}	18.22 ^{a}
	Sun-dried	86.21 ^{b}	6.07 ^{a}	8.51 ^{b}	25.27 ^{a}	4.48 ^{b}	13.67 ^{b}
	Std. Dev.	1.84	0.67	1.69	2.79	1.48	3.22
Lemon grass	Oven-dried	91.53 ^{b}	9.81 ^{a}	10.26 ^{a}	25.84 ^{a}	3.87 ^{a}	17.60 ^{a}
	Sun-dried	91.75 ^{a}	7.96 ^{b}	10.40 ^{a}	25.98 ^{a}	3.35 ^{b}	12.30 ^{b}
	Std. Dev.	0.16	1.31	0.10	0.10	0.37	3.75
Common fennel	Oven-dried	94.12 ^{a}	13.19 ^{a}	24.68 ^{b}	10.56 ^{a}	3.35 ^{a}	16.52 ^{a}
	Sun-dried	90.90 ^{b}	13.16 ^{a}	27.08 ^{a}	10.84 ^{a}	2.68 ^{a}	13.33 ^{a}
	Std. Dev.	2.28	0.02	1.70	0.20	0.47	2.26
Mint	Oven-dried	92.20 ^{a}	11.44 ^{a}	27.47 ^{a}	9.67 ^{a}	4.99 ^{a}	16.77 ^{a}
	Sun-dried	91.56 ^{a}	11.41 ^{a}	24.47 ^{a}	9.62 ^{a}	3.80 ^{b}	18.84 ^{a}
	Std. Dev.	0.46	0.02	2.12	0.04	0.84	1.46
Basil	Oven-dried	94.50 ^{a}	16.13 ^{a}	25.49 ^{a}	11.28 ^{a}	3.78 ^{b}	11.97 ^{b}
	Sun-dried	88.97 ^{b}	9.15 ^{b}	16.09 ^{b}	7.84 ^{b}	7.27 ^{a}	30.93 ^{a}
	Std. Dev.	3.91	4.94	6.65	2.43	2.47	13.41

[1] For each herb and each component, mean values bearing dissimilar superscript lowercase letters (a,b) are significantly different according to Tukey's test ($p < 0.05$) and $n = 2$.

3.3. Microbiological Characteristics of Oven- and Sun-Dried Herbs

As shown in Table 3, the levels of mold and yeast in most dried herbs were within the intermediate quality established by the Brazilian National Health Surveillance Agency [27] for fruits and vegetables. However, the amounts of these microorganisms were well above

the limit in the oven- and sun-dried basil and in the common fennel processed in the solar dryer. Contamination of herbs may have occurred during the handling of the plant material when the leaves were distributed in the drying trays or during the packaging of the dried material prior to analyses.

Table 3. Microbial contamination of oven- and sun-dried herbs.

Drying Process	Microbial Counts				
	Rosemary	**Lemon Grass**	**Common Fennel**	**Mint**	**Basil**
Mold and yeast (CFU/g) [1]					
Oven-dried	0.40×10^3	1.1×10^3	1.3×10^3	0.70×10^3	5.6×10^3
Sun-dried	0.30×10^3	2.0×10^3	1.6×10^4	2.0×10^3	9.6×10^4
Staphylococcus aureus (CFU/g)					
Oven-dried	0	0	3.66×10^3	0	5.99×10^2
Sun-dried	0	0	6.66×10^3	0	0
Total coliforms (MPN/g) [2]					
Oven-dried	11.5	16.0	240	23.0	240
Sun-dried	0	151.5	1100	1100	16.05
Fecal coliforms (MPN/g)					
Oven-dried	0	0	13.3	0	0
Sun-dried	0	79.55	1.8	6.35	4.55

[1] CFU, colony-forming unit. [2] MPN, most probable number.

Contamination with *S. aureus* was detected in the oven- and sun-dried common fennel and in oven-dried basil, and the amounts of bacteria exceeded the upper limit of 10^2 CFU/g coagulase-positive staphylococci permitted by the Brazilian National Health Surveillance Agency [28]. Nevertheless, the level of contamination in basil was an order of magnitude lower than that observed in common fennel. No sulfite-reducing clostridia or *Salmonella* sp. were detected in any herb samples, irrespective of the drying method employed.

Total coliforms were present in all samples of dehydrated herbs, irrespective of the drying process applied, with the single exception of sun-dried rosemary. Fecal coliforms were absent from oven-dried rosemary, lemon grass, mint, and basil and from sun-dried rosemary samples, while the other dried herbs presented various degrees of contamination with oven-dried common fennel and sun-dried lemon grass exhibiting levels above the limit of 10 MPN/g permitted by legislation [28]. The contamination of dried basil was surprising since this plant appears to have bioactive properties against *E. coli*, *Salmonella typhimurium*, *S. aureus*, *Yersinia enterocolitica*, *Enterococcus faecalis*, and *Bacillus cereus* [29]. It is evident that hygiene should be ensured during the manipulation of fresh and dried plants, together with the use of alternative methods of decontamination.

3.4. Color Characteristics of Oven- and Sun-Dried Herbs

As shown by the L* values (Table 4), the brightness of all dehydrated herbs tended towards the dark end of the spectrum, with the lightest samples being those of sun-dried rosemary and the darkest being those of sun-dried common fennel. With the exception of lemon grass, the drying process significantly affected the lightness of the herbs.

Table 4. Values of the Commission Internationale de l'Eclairage color space parameters L*a*b* of oven- and sun-dried herbs.

Herbs	Drying Process	Color Space Parameters [1]		
		L* [2]	a* [3]	b* [4]
Rosemary	Oven-dried	42.31 [b]	0.90 [a]	14.97 [b]
	Sun-dried	49.12 [a]	0.85 [a]	16.47 [a]
	Std. Dev.	4.82	0.04	1.06
Lemon grass	Oven-dried	45.42 [a]	−4.32 [b]	29.17 [a]
	Sun-dried	45.30 [a]	−5.20 [a]	23.98 [b]
	Std. Dev.	0.09	0.62	3.67
Common fennel	Oven-dried	30.91 [a]	−3.92 [b]	13.13 [b]
	Sun-dried	29.37 [b]	−4.45 [a]	13.26 [a]
	Std. Dev.	1.09	0.38	0.09
Mint	Oven-dried	33.30 [b]	−2.41 [b]	15.27 [b]
	Sun-dried	35.65 [a]	−3.57 [a]	16.62 [a]
	Std. Dev.	1.66	0.82	0.96
Basil	Oven-dried	34.69 [b]	−0.79 [b]	14.46 [b]
	Sun-dried	42.60 [a]	−3.23 [a]	20.09 [a]
	Std. Dev.	5.59	1.73	3.98

[1] For each herb and each parameter, mean values bearing dissimilar superscript lowercase letters (a,b) are significantly different according to Tukey's test ($p < 0.05$) and $n = 2$. [2] Lightness from black (0) to white (100). [3] Band green (−) to red (+). [4] Band blue (−) to yellow (+).

The recorded values of a* indicated that all dried herb samples, except for those of rosemary, tended towards the green rather than the red end of the spectrum, and this was particularly the case for herbs that had been sun-dried. It is likely that the higher temperature of the oven (50 °C) compared with that of the solar dryer might have led to the degradation of some photosynthetic pigments. The b* values showed that all dried herb samples tended towards the yellow rather than the blue end of the spectrum. Oven-dried lemon grass exhibited the largest yellow component in comparison with the other herbs, probably because of the extended time/temperature interaction, since this herb took the longest time to reach equilibrium moisture in the oven (Figure 2a). Regarding the other herbs, the highest b* values were observed in sun-dried samples, a finding that can be explained by the protracted time required to reach the equilibrium moisture under the solar dryer.

3.5. Active Principles in of Oven- and Sun-Dried Herbs

Extracts of all dried herb samples exhibited antioxidant properties, although the recorded activities were lower than those of the positive controls at the concentrations assessed (Table 5). The antioxidant activities of oven-dried herbs ranged from 19.18 to 71.55% and increased in the order common fennel < lemon grass < mint < basil < rosemary, while the activities of sun-dried samples varied from 17.73 to 58.27% and increased in the order basil < common fennel < lemon grass < mint < rosemary. These results may be compared with those of Lee and Shibamoto [30], who reported that extracts of rosemary and basil presented the highest antioxidant activities among the various herbs tested. In the present study, herbs that had been oven-dried exhibited the highest antioxidant activities, with the exception of common fennel. The process of solar drying was much slower than that of oven drying, and the increased process time favored the loss of active principles even though the average drying temperature in the solar dryer (31.6 °C) was lower than that of the oven (50 °C). Whilst oven-dried samples were more active than their sun-dried counterparts, the differences between the two values were not statistically significant except in the case of basil. This may have been caused by environmental factors such as

temperature, humidity, soil pH, winds, and air pollution, among others, since all these factors are known to be responsible for fluctuations in plant secondary metabolites [26].

Table 5. Antioxidant activity and concentration of antioxidant compounds in the herbs.

Drying Process	Herbs					Positive Controls	
	Rosemary	Lemon Grass	Common Fennel	Mint	Basil	Butylated Hydroxy-toluene	α-Tocopherol
Antioxidant activity (%)							
Oven-dried	71.55 [a]	55.46 [a]	19.18 [a]	56.06 [a]	60.54 [a]	89.53 [a]	90.23 [a]
Sun-dried	58.27 [a]	38.99 [a]	22.41 [a]	42.98 [a]	17.73 [b]	90.56 [a]	87.75 [a]
Std. Dev.	9.18	12.00	13.10	9.65	30.13	-	-
Total phenolics (mg gallic acid/g extract)							
Oven-dried	165.68 [b]	93.04 [a]	31.43 [b]	115.27 [a]	91.46 [b]	-	-
Sun-dried	182.20 [a]	95.56 [a]	49.31 [a]	126.20 [a]	149.28 [a]	-	-
Std. Dev.	12.68	9.36	12.64	7.73	40.89		
Total flavonoids (mg quercetin/g extract)							
Oven-dried	9.96 [a]	13.79 [a]	5.15 [a]	20.00 [a]	0.75 [b]	-	-
Sun-dried	9.45 [a]	13.67 [a]	5.00 [a]	20.78 [a]	13.02 [a]	-	-
Std. Dev.	0.36	0.08	0.11	0.55	8.68		

For each herb and each activity/concentration, mean values bearing dissimilar superscript lowercase letters (a,b) are significantly different according to Tukey's test ($p < 0.05$) and $n = 2$.

The antioxidant activities of oven-dried herbs showed a strong and positive correlation (r = 0.92) with the concentrations of total phenolics in their respective extracts, whereas in sun-dried samples, the correlation was moderate and positive (r = 0.53). Oven- and sun-dried samples of rosemary exhibited the highest antioxidant activities and contained the highest amounts of phenolics. Extracts of rosemary are recognized for their significant antioxidant properties, and these have been attributed to the high content of carnosic acid, carnosol, and rosmarinic acid [31,32].

Unlike total phenolics, the concentrations of total flavonoids in extracts of dried herbs were weekly and positively correlated (r = 0.22) with the antioxidant activities of the herbs regardless of the drying process. Thus, while the concentrations of flavonoids in dried rosemary extracts were not particularly high, the antioxidant activities of the oven- and sun-dried samples were the highest among the studied herbs. It is, therefore, reasonable to assume that the strong antioxidant activity of dried rosemary is due only to a limited extent to the presence of flavonoids. In contrast, the concentration of flavonoids in oven-dried basil was 17-fold lower than that in the sun-dried herb, and yet the antioxidant activity of the oven-dried herb was some 3.5 times higher. Extracts of common fennel contained the lowest concentrations of phenolics and flavonoids among all herbs tested and, consequently, the lowest antioxidant activity.

Besides acting as antioxidants, phenolic compounds are beneficial to health in various other ways. For example, phenolics possess anti-bacterial, anti-fungal, and anti-cancer properties and also help to prevent cardiovascular diseases [33].

4. Conclusions

Oven drying was influenced by the temperature applied, while sun drying was affected by both temperature and relative humidity, and such conditions determined the differential characteristics of the dried herbs so produced. All herbs presented much faster drying rates in the forced-air oven than in the solar dryer. The highest and lowest oven drying rates were observed for common fennel and lemon grass, respectively, while in the solar dryer, the highest drying rate was observed for lemon grass and the lowest for rosemary. In general, the centesimal compositions and colors of oven-dried and sun-dried herbs were significantly different, as were the levels of microbial contamination assessed in

the samples. Mint appeared to be the least affected by changes in drying conditions in that only color and amount of reducing sugars showed significant differences in the two drying processes. The antioxidant properties of the dried herbs, which were due primarily to the presence of total phenolics, were mainly similar regardless of the drying process employed. Neither drying process was sufficient to impede the growth of pathogens, signifying that additional strategies must be employed to ensure adequate decontamination and preservation of dried herbs, such as careful handling and storage of herbs. Taken together, the results obtained support our original hypothesis that the use of a solar dryer rather than a forced-air oven would affect the final quality of an herbal product. Our study is, therefore, important because it demonstrates that the process of drying can alter the quality of an herbal product, implying that standardization of this post-harvesting step is essential to ensure the consistency of quality, efficacy, and safety of a phytopharmaceutical.

Author Contributions: A.P.M.: analyses, methodology, writing—original draft preparation. S.M.A.: methodology, formal analysis, investigation. A.F.S.: data interpretation. M.P.M.: design the investigation, visualization. I.G.V.: review and editing, conceptualization. A.L.G.F.: study design, writing, supervision. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Fundação de Estudos Agrários Luiz de Queiroz and the São Paulo Research Foundation (FAPESP).

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

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

References

1. World Health Organization. WHO | World Health Organization. 2020. Available online: https://www.who.int/ (accessed on 23 February 2022).
2. Ekor, M. The growing use of herbal medicines: Issues relating to adverse reactions and challenges in monitoring safety. *Front. Pharmacol.* **2014**, *4*, 177. [CrossRef]
3. World Health Organization. *WHO Guidelines on Good Agricultural and Collection Practices [GACP] for Medicinal Plants*; World Health Organization: Geneva, Switzerland, 2003.
4. Morgan, R. *Enciclopédia Das Ervas e Plantas Medicinais*, 9th ed.; Editora Hemus: São Paulo, Brazil, 2003.
5. Apolinário, P. Rede Sociotécnica de Inovação em Plantas Medicinais e Fitoterápicos: Do Coletivo de Mulheres no Assentamento Pirituba ao SUS Itapeva-SP. Master's Thesis, Universidade Federal de São Carlos, São Paulo, Brazil, 2021.
6. De Figueredo, C.A.; Gurgel, I.G.D.; Gurgel Junior, G.D. A Política Nacional de Plantas Medicinais e Fitoterápicos: Construção, perspectivas e desafios. *Physis* **2014**, *24*, 381–400. [CrossRef]
7. Chen, B.; Xu, M. Natural Antioxidants in Foods. In *Encyclopedia of Food Chemistry*; Varelis, P., Melton, L., Shahidi, F., Eds.; Elsevier: Amsterdam, The Netherlands, 2019.
8. Brainina, K.; Stozhko, N.; Bukharinova, M.; Khamzina, E.; Vidrevich, M. Potentiometric method of plant microsuspensions antioxidant activity determination. *Food Chem.* **2019**, *278*, 653–658. [CrossRef] [PubMed]
9. Milevskaya, V.V.; Prasad, S.; Temerdashev, Z.A. Extraction and chromatographic determination of phenolic compounds from medicinal herbs in the Lamiaceae and Hypericaceae families: A review. *Microchem. J.* **2019**, *145*, 1036–1049. [CrossRef]
10. Nazarenko, D.V.; Kharyuk, P.V.; Oseledets, I.V.; Rodin, I.A.; Shpigun, O.A. Machine learning for LC–MS medicinal plants identification. *Chemom. Intell. Lab. Syst.* **2016**, *156*, 174–180. [CrossRef]
11. Pedrete, T.A.; Hauser-Davis, R.A.; Moreira, J.C. Proteomic characterization of medicinal plants used in the treatment of diabetes. *Int. J. Biol. Macromol.* **2019**, *140*, 294–302. [CrossRef]
12. Steinhoff, B. Review: Quality of herbal medicinal products: State of the art of purity assessment. *Phytomedicine* **2019**, *60*, 153003. [CrossRef]
13. Geankoplis, C.J. *Transport Processes and Separation Process Principles*; Prentice Hall: New York, NY, USA, 2003.
14. Association of Official Analytical Chemists. *Official Methods of Analysis of the Association of Official Analytical Chemists*; Association of Official Analytical Chemists: Arlington, VA, USA, 1990.
15. Quansah, J.K.; Gazula, H.; Holland, R.; Scherm, H.; Li, C.; Takeda, F.; Chen, J. Microbial quality of blueberries for the fresh market. *Food Control.* **2019**, *100*, 92–96. [CrossRef]
16. Picoli, S.U.; Bessa, M.C.; Castagna, S.M.F.; Gottardi, C.P.T.; Schmidt, V.; Cardoso, M. Enumeration of coliforms, *Staphylococcus aureus* and aerobic mesofilic bacteria throughout the manufacture process of a goat unripened cheese produced in a dairy plant. *Food Sci. Technol.* **2006**, *26*, 64–69. [CrossRef]

17. Prevost, S.; Cayol, J.-L.; Zuber, F.; Tholozan, J.-L.; Remize, F. Characterization of clostridial species and sulfite-reducing anaerobes isolated from foie gras with respect to microbial quality and safety. *Food Control.* **2013**, *32*, 222–227. [CrossRef]
18. Ministério da Saúde. *Manual Técnico de Diagnóstico Laboratorial da Salmonella spp.*; Secretaria de Vigilância em Saúde. Departamento de Apoio à Gestão de Vigilância em Saúde: Brasília, Brazil, 2011. Available online: https://portalarquivos2.saude.gov.br/images/pdf/2014/dezembro/15/manual-diagnostico-salmonella-spp-web.pdf (accessed on 3 March 2022).
19. Ministério da Agricultura. *Métodos Analíticos Oficiais para Controle de Produtos de Origem Animal e seus Ingredientes. II-Métodos Microbiológicos*; Secretaria Nacional de Defesa Agropecuária. Laboratório Nacional de Referência Animal: Brasília, Brazil, 2018. Available online: https://www.gov.br/agricultura/pt-br/assuntos/lfda/legislacao-metodos-da-rede-lfda/poa/metodos_oficiais_para_analise_de_produtos_de_origem_animal-_1a_ed-_2022_assinado.pdf (accessed on 17 May 2023).
20. Singleton, V.L.; Orthofer, R.; Lamuela-Raventós, R.M. Analysis of total phenols and other oxidation substrates and antioxidants by means of folin-ciocalteu reagent. *Methods Enzymol.* **1999**, *299*, 152–178. [CrossRef]
21. Park, J.C.; Lee, J.H.; Choi, J.S. A flavone diglycoside from *Cirsium japonicum* var. ussuriense. *Phytochemistry* **1995**, *39*, 261–262. [CrossRef]
22. Emmons, C.L.; Peterson, D.M.; Paul, G.L. Antioxidant Capacity of Oat (*Avena sativa* L.) Extracts. 2. In Vitro Antioxidant Activity and Contents of Phenolic and Tocol Antioxidants. *J. Agric. Food Chem.* **1999**, *47*, 4894–4898. [CrossRef] [PubMed]
23. Doymaz, I. Thin-layer drying behaviour of mint leaves. *J. Food Eng.* **2006**, *74*, 370–375. [CrossRef]
24. Park, K.J.; Vohnikova, Z.; Brod, F.P.R. Evaluation of drying parameters and desorption isotherms of garden mint leaves (*Mentha crispa* L.). *J. Food Eng.* **2002**, *51*, 193–199. [CrossRef]
25. Akpinar, E.K. Mathematical modelling of thin layer drying process under open sun of some aromatic plants. *J. Food Eng.* **2006**, *77*, 864–870. [CrossRef]
26. Yang, L.; Wen, K.-S.; Ruan, X.; Zhao, Y.-X.; Wei, F.; Wang, Q. Response of Plant Secondary Metabolites to Environmental Factors. *Molecules* **2018**, *23*, 762. [CrossRef]
27. Agência Nacional de Vigilância Sanitária. *Instrução Normativa n° 60 de 23 de Dezembro de 2019. Estabelece as Listas de Padrões Microbiológicos para Alimentos*; ANVISA: Brasília, Brazil, 2019. Available online: https://www.in.gov.br/en/web/dou/-/instrucao-normativa-n-60-de-23-de-dezembro-de-2019-235332356 (accessed on 3 March 2022).
28. Agência Nacional de Vigilância Sanitária. *Resolução-RDC N° 12, de 2 de Janeiro de 2001. Regulamento Técnico Sobre Padrões Microbiológicos Para Alimentos*; ANVISA: Brasília, Brazil, 2001. Available online: http://portal.anvisa.gov.br/documents/33880/2568070/RDC_12_2001.pdf/15ffddf6-3767-4527-bfac-740a0400829b (accessed on 3 March 2022).
29. Nascimento, G.G.F.; Locatelli, J.; Freitas, P.C.; Silva, G.L. Antibacterial activity of plant extracts and phytochemicals on antibiotic-resistant bacteria. *Braz. J. Microbiol.* **2000**, *31*, 247–256. [CrossRef]
30. Lee, K.-G.; Shibamoto, T. Determination of Antioxidant Potential of Volatile Extracts Isolated from Various Herbs and Spices. *J. Agric. Food Chem.* **2002**, *50*, 4947–4952. [CrossRef]
31. Penuelas, J.; Munnebosch, S. Isoprenoids: An evolutionary pool for photoprotection. *Trends Plant Sci.* **2005**, *10*, 166–169. [CrossRef]
32. Wellwood, C.R.L.; Cole, R.A. Relevance of Carnosic Acid Concentrations to the Selection of Rosemary, *Rosmarinus officinalis* (L.), Accessions for Optimization of Antioxidant Yield. *J. Agric. Food Chem.* **2004**, *52*, 6101–6107. [CrossRef] [PubMed]
33. Morton, L.W.; Caccetta, R.A.-A.; Puddey, I.B.; Croft, K.D. Chemistry and Biological Effects of Dietary Phenolic Compounds: Relevance to Cardiovascular Disease. *Clin. Exp. Pharmacol. Physiol.* **2000**, *27*, 152–159. [CrossRef] [PubMed]