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The aim of the study was to determine the effect of the method and temperature of the sublimation, vacuum and convective drying process on the kinetics of dehydration of the Moldovan dragonhead leaves, as well as on the physical and chemical properties, such as color coordinates, total content of phenolic compounds, antioxidant capacity, total monomeric anthocyanins content, total flavonoids content and content of essential oils. It was shown that the kinetics of the sublimation and vacuum drying process are best described by the logarithmic and Page models. Sublimation and vacuum drying were characterized by comparable process times. An increase in drying temperature caused a decrease in the content of phenolic compounds and antioxidant activity of the dried material and a significant change in the composition of essential oils. The lowest brightness of the dried material and the lowest color saturation was found after sublimation drying at 20 °C. The highest dried material quality was found in case of convective and sublimation drying at 20 °C.

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Drying Characteristics of *Dracocephalum moldavica* Leaves: Drying Kinetics and Physicochemical Properties

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Abstract: The aim of the study was to determine the effect of the method and temperature of the sublimation, vacuum and convective drying process on the kinetics of dehydration of the Moldovan dragonhead leaves, as well as on the physical and chemical properties, such as color coordinates, total content of phenolic compounds, antioxidant capacity, total monomeric anthocyanins content, total flavonoids content and content of essential oils. It was shown that the kinetics of the sublimation and vacuum drying process are best described by the logarithmic and Page models. Sublimation and vacuum drying were characterized by comparable process times. An increase in drying temperature caused a decrease in the content of phenolic compounds and antioxidant activity of the dried material and a significant change in the composition of essential oils. The lowest brightness of the dried material and the lowest color saturation was found after sublimation drying at 20 °C. The highest dried material quality was found in case of convective and sublimation drying at 20 °C.

Keywords: Moldavian dragonhead; drying; flavonoids content; antioxidant capacity; essential oils; color changes

1. Introduction

The Moldavian dragonhead (*Dracocephalum moldavica*) is an annual plant growing up to about 80 cm. There are two varieties of this plant, with white and blue flowers, and it belongs to the Lamiaceae family. Under natural conditions, the plant is found in China and Mongolia, central Asia and central and eastern Europe [1,2].

The dried leaves of the Moldavian dragonhead have a lemon aroma and are used in natural medicine to prepare infusions due to their antiseptic, antibacterial, antioxidant, calming and appetite-stimulating properties [3]. The herb of the Moldavian dragonhead contains mainly terpenoids: neral, geraniol, geranyl acetate and flavonoids, iridoids as well as tannins [4,5]. The composition of the essential oil depends mainly on the climate in which the Moldavian dragonhead was grown. The oil obtained from cultivation in temperate climates contains higher amounts of alcoholic monoterpene derivatives, and from cultivation in warmer climates it contains higher amounts of aldehyde and ester monoterpene derivatives [6]. The oil and extracts from the dried dragonhead leaves are used in the food industry [7,8]. Dried leaves of Moldavian dragonhead can be used as tea [9], as well as a functional additive for bread [10] and snacks production [11]. A high moisture content in the leaves dramatically reduces it shelf life and decreases its quality. Thus, the fast and proper preservation of bioactive compounds through the processing of dragonhead leaves is a very important issue.One of the main methods of food and medicinal plants preservation is drying, which allows for long-term storage of dried food by reducing water activity and the related reduction of microbial development, as well as mass reduction and elimination of seasonal availability of raw materials. However, drying of food is associated with a number of adverse changes occurring during the process. These changes are mainly due to the use of increased drying temperatures required to achievethe optimum water content [12,13].

For most plant raw materials, due to their health-promoting properties, the aim is to limit the adverse physicochemical changes occurring during drying. A compromise is needed between the drying kinetics, the intensification of which makes it possible to obtain more dried material, the energy intensity of the process, which makes it possible to reduce the company's costs, and the change in the physicochemical properties of the dried product, which is the main indicator of consumer preferences. In order to reconcile these contradictory relations, it is necessary to select the optimal method and parameters of drying for a given raw material. Attempts are also made to apply variable drying conditions (during the process), the use of intermittent drying and to combine different drying methods during one technological process [14,15]. Plants containing essential oils were added to food to improve taste and were also valued for their aromatic properties. Nowadays, plant essential oils are increasingly used in medicine as they have antibacterial, antiviral, anticancer, antifungal, spasmolytic andhepatotropic properties [16–18]. Essential oils and other bioactive compounds contained in the plant raw materials are impermanent and, as a result of drying, their amount is reduced; there are also reactions consisting of the formation of new components from the existing oil components [19,20]. Guo et al. [21] studied the drying process of lotus leaves and they found that an air-drying temperature between 55 and 65 °C is the most adequate to keep the higher antioxidant activity in leaves. Yap et al. [22] showed that an air-drying temperature higher than 60 °C decreased the total phenolics content in papaya leaves. Moreover, they proved that freeze-drying is the best method to preserve bioactive compounds in papaya leaves. A similar tendency was found by Choo et al. [23] during drying of *Murraya koenigii* leaves. They also found that an air-drying temperature higher than 40 °C significantly decreases antioxidant activity. De Suza et al. [24] showed that an air-drying temperature higher than 50 °C causes significant decreases in the content of the essential oil of clove basil leaves. However, other authors found that the highest essential oil content in dry *Piper umbellatum* L. leaves is obtained when the air-drying temperature does not exceed 40 °C [25]. On the basis of the presented data it can be concluded that the quantity and quality of oils and other bioactive compounds are most affected by the drying method and drying temperature.

Therefore, this study aimed to determine the effect of the temperature of the sublimation, vacuum and convective drying process on the kinetics of drying of the Moldavian dragonhead leaves, as well as onthe selected physical and chemical properties of the dried material (color change, polyphenols content, antioxidant capacity, total monomeric anthocyanins content, total flavonoids content and essential oil content).

2. Materials and Methods

2.1. Material

The Moldavian dragonhead (*Dracocephalum moldavica*) with white flowers came from the experimental station of the University of Life Sciences in Lublin (51°34′ N, 23°02′ E), Poland. The dragonhead leaves were collected from three plots (50 m^2 each). The tillage system for this plant was described by Wójtowicz et al. [11]. Plants of the Moldavian dragonhead were cut off from three plots between 12 and 14 July 2018 (at the beginning of the flowering phase).After harvesting, leaves weremanually collected, placed in paper bags and stored at 10 °C for 3 h before convection drying and vacuum drying.The leaves were dried whole, and before sublimation drying they were frozen at −30 °C for 24 h. The moisture content of the fresh leaves of the Moldavian dragonhead was determined by weight method, drying them under vacuum at 95 °C ± 0.5 °C to obtain a constant weight. The moisture content of the leaves of the Moldavian dragonhead was 83.24% ± 0.35% (wet basis). Each method of drying was performed individually for leaves from each plot. In order to standardize the sample, dried leaves from each plot were mixed.

2.2. Drying Method

The convection drying process was carried out in a laboratory convection dryer (Promis Tech, Wrocław, Poland). Drying was carried out at a drying air temperature of 40 °C and 60 °C and a constant air flow velocity of 0.5 $m \cdot s^{-1}$. Registration of changes in the mass of the material during drying was provided by a laboratory electronic scale, type B3 (AXIS, Gdańsk, Poland) with an accuracy of ±0.1 g, connected to a computer recording its indications. The drying process led to a final material moisture content of 10%.

The sublimation and vacuum drying processes were carried out in an ALPHA 1–4 (Martin Christ Company, Osterode am Harz, Germany) laboratory vacuum dryer. The device consists of a drying chamber, vacuum pump, ice capacitor, heating system consisting of 5 shelves placed on a rack and a control and measurement system. The device is equipped with a weighing system that records, in a continuous manner, changes in weight during the process (accuracy of weight ±0.1 g). The heat required for the phase transition is delivered to the material by contact. The heating panel temperature control is performed by means of a temperature sensor placed inside one of the lyophilizer panels [26]. The sublimation drying process was carried out for heating panel temperatures of 20 °C, 40 °C and 60 °C with a constant pressure value of 63 Pa. Vacuum drying was carried out for heating panel temperatures of 20 °C, 40 °C and 60 °C with a constant pressure value of 2000 Pa (in case of vacuum drying, the material was not previously frozen). Sublimation and vacuum drying were carried out to obtain a constant dried material humidity of 10%.We did not carry out convection drying at 20 °C, because this kind of drying at such low temperature is ineffective, takes a long time, can lead to the development of microorganisms and often does not allow to achieve an adequate level of moisture content after drying. The drying temperature was selected on the basis of the literature data mentioned in the introduction [21–25].

2.3. Modeling of Drying Curves

The drying kinetics is presented as a change in reduced water content (*MR*) as a function of drying time:

$$MR = \frac{u_t - u_r}{u_0 - u_r}, \tag{1}$$

where u_t is the water content in the course of drying (kg H_2O/kg DM), u_0 is the initial water content (kg $H_2O \cdot$kg DM^{-1})and u_r is the equilibrium water content (kg $H_2O \cdot$kg DM^{-1}). The equilibrium water content after sublimation and vacuum drying is very low, so the equilibrium water content (u_r) is assumed to be 0 over the entire measuring range. Such simplification has little effect on drying kinetics [27–29].

The most frequent models in the literature were used to describe the curves of sublimation, vacuum and convective drying. The model equations are presented in Table 1.

Table 1. Equations applied to the drying curves.

Model Number	Model Name	Model Equation	References
1	Newton [1]	$MR = \exp(-k \cdot \tau)$	[30]
2	Page	$MR = \exp(-k \cdot \tau^n)$	[31]
3	Henderson and Pabis	$MR = a \cdot \exp(-k \cdot \tau)$	[32]
4	Logarithmic	$MR = a \cdot \exp(-k \cdot \tau) + b$	[33]
5	Wang and Singh	$MR = 1 + a \cdot \tau + b \cdot \tau^2$	[34]

[1] *k*—Drying coefficient (min^{-1}); *a*, *b*—Coefficients of the equations; *n*—Exponent; *τ*—Time (min).

2.4. Measurement of Color Coordinates

Color measurement was performed using the reflection method with an X-Rite 8200 spherical spectrophotometer with a measuring hole of 12.7 mm in diameter. A D_{65} light source and a standard colorimetric observer with a 10° field of view were used. Before each measurement, the instrument was calibrated using a white standard.

The color coordinates were determined in the *CIEL*a*b** system. The measurement of color in this system is based on the numerical determination of the three coordinates *L**, *a**, *b**, where *L** stands for color brightness and ranges from 0 for a perfectly black body to 100 for a perfectly white body. The coordinate *a** indicates the color change from green *(−a*)* to red *(+a*)*, and *b** indicates the color change from blue *(−b*)* to yellow *(+b*)*.

On the basis of the designated color coordinates, the total change in color (Δ*E*) in relation to the raw material was determined, as recorded in the cylindrical coordinates for the color saturation *(C)* and color shade *(HU)* of the dried material [35].

$$C = \sqrt{(a^*)^2 + (b^*)^2}, \tag{2}$$

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}, \tag{3}$$

$$\Delta L = L^* - L^*_{0,} \Delta a = a^* - a^*_{0,} \Delta b = b^* - b^*_{0,}, \tag{4}$$

$$HU = \tan^{-1}\frac{b}{a}, \tag{5}$$

where L_0*, a_0* and b_0* are the color parameters of the fresh sample.

2.5. Total Phenolic Compounds Content

The content of total phenolic compounds (*TPC*) was determined by the Folin–Ciocalteu method [36]. A 0.5 mL extract sample was mixed with 0.5 mL H_2O, 2 mL Folin reagent (1:5 H_2O) and, after 3 min, with 10 mL 10% Na_2CO_3. After 30 min, the absorbance of the mixed samples was measured at 725 nm. The measurement was carried out on a UV spectrophotometer Mini 1240 (Shimadzu, Japan). The content of the total phenolic compounds was expressed as gallic acid equivalent per gram of dry matter.

2.6. Antiradical Activity

The ability to neutralize free radicals against ABTS (2,2′-azinobis (3-ethylbenzothiazoline-6-sulfonate)) was carried out by the method developed by Re et al. [37]. The ability to neutralize free radicals against DPPH (2,2-diphenyl-1picyclohydrazil) was carried out according to Brand-Williams et al. [38]. The decrease in absorbance was measured quantitatively on the spectrophotometer at 734 nm for ABTS and 517 nm for DPPH.

The ability to neutralize free radicals of ABTS and DPPH was expressed as the EC_{50} index—the dry matter concentration (mg·mL^{-1}) causing a 50% decrease in the initial ABTS or DPPH radical concentration.

2.7. Total Flavonoids Content

Total flavonoids were estimated according to the method described by Bahorun et al. [39]. One milliliter of sample was mixed with 1 mL 2% $AlCl_3 \cdot 6H_2O$. After 10 min, absorbance at 430 nm was measured. The total flavonoids content was expresses as quercetin equivalent (QE) in milligrams per dry weight (DW).

2.8. Total Monomeric Anthocyanins

Total monomeric anthocyanins (*TAC*) were quantified by the pH-differential method according to Giusti and Wrolstad [40]. Extracts of cranberry powders were diluted with two buffer solutions at pH 1 and 4.5. Cy-3-G (MW = 449.2 g·mol^{-1}) was used as a standard with a molar absorptivity coefficient of 26900. Results were expressed as mg of cyanidin-3-galactoside equivalent as mg of Cy-3-G per g DM.

2.9. Quantity and Composition of Essential Oils

The volume of essential oil was determined by steam distillation in the Dering apparatus. The 20 g of crushed material was placed in a distillation flask containing 400 mL of distilled water. The distillation process was carried out for 3 h from the start of boiling. The resulting volume of oil was converted into 100 g of dry matter. The qualitative composition and percentage content of the essential oil components was determined by GC/MS method, using the Varian Chrompack CP-3800 gas chromatograph with Varian 4000 MS/MS mass detector and flame ionization detector (FID). The VF-5 ms column (equivalent to DB-5) was used. The carrier gas was helium with a constant flow through the column of 0.5 mL·min^{-1}. The column temperature program was as follows: 50 °C for 1 min, increased to 250 °C at 4 °Cmin^{-1}, and then 250 °C for 10 min. Doser: 250 °C, split 1:50. A total of 1 µL of solution was dosed (10 µL of sample in 1000 µL of hexane). The recorded range was 40–1000 m·z^{-1}, and the scanning rate 0.8 sec·skan^{-1}. Kovats retention indices (non-isothermal Kovats retention index) were determined on the basis of the C10–C40 alkanes series, using the NIST Library and HP Chemstation spectral libraries.

2.10. Statistical Analysis of the Results

One- and two-factor analysis of variance was performed. Tukey's test was used to determine the significance of differences between the means. Each drying test was performed in triplicate and all analyses were performed in 5 repetitions. The analysis of drying kinetics regression was performed by non-linear estimation using the least squares method, determining the determination factor and the root mean square error (*RMSE*) as well as chi-squared test values (χ^2). The mean root square error (*RMSE*) and chi-squared test values (χ^2) were determined from the relationship

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N}\left(MR_{i,p} - MR_{i,e}\right)^2}{N}}, \quad (6)$$

$$\chi^2 = \frac{\sum_{i=1}^{N}\left(MR_{i,p} - MR_{i,e}\right)^2}{N - n}, \quad (7)$$

where $MR_{i,p}$—predicted value of reduced water content; $MR_{i,e}$—experimental value of reduced water content; N—number of measurements; and n—number of parameters in the model equation.

Statistica 13 from StatSoft was used for statistical analysis. All calculations were performed with the significance level α = 0.05. This level of α is used in many biological studies.

3. Results and Discussion

3.1. Drying Kinetics

Changes in the reduced water content (*MR*) as a function of the duration of sublimation, vacuum and convective drying of the dragonhead leaves is shown in Figures 1–3. On the basis of the conducted tests, it can be concluded that with the increase in drying temperature, the duration of the process is reduced—for all the analyzed drying methods. Increasing the temperature from 20 °C to 60 °C resulted in shortening the drying time by about 34% (sublimation drying) and by 32% (vacuum drying). An increase in temperature from 40 °C to 60 °C shortened the convective drying time by about 45%. At a given temperature level, vacuum drying was shorter than sublimation drying. However, the differences were small and amounted to about 20 min. The shortest time was observed in the case of convective drying: at 60 °C, the time was 160 min.

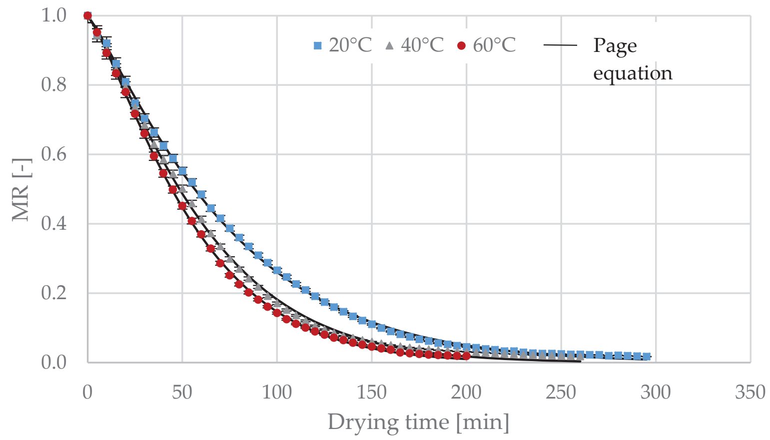

Figure 1. Drying curves of the freeze-drying of Moldavian dragonhead.

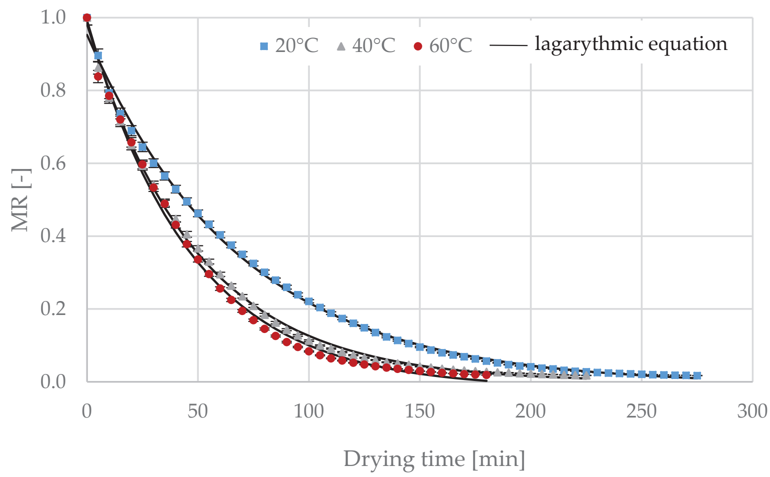

Figure 2. Drying curves of the vacuum drying of Moldavian dragonhead.

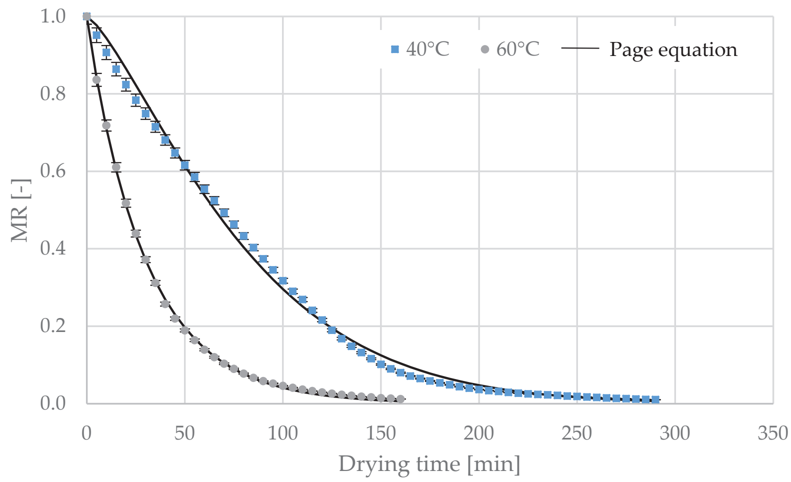

Figure 3. Drying curves of the air drying of Moldavian dragonhead.

The results of the regression analysis of the five analyzed models used to describe the kinetics of sublimation, vacuum and convective drying are presented in Tables 2–4. It can be seen that a good match of the experimental data was obtained for each of the analyzed models. The value of the coefficient of determination (R^2) of the equations ranged from 0.932 to 0.999. The square root from the mean square error (*RMSE*) and the value of the reduced test (χ^2) had small values ranging from 0.0033–0.0678 and 0.00001–0.00481, respectively. In case of vacuum drying, the best match between the analyzed models and experimental data was obtained using a logarithmic model (R^2—above 0.997; *RMSE*—below 0.0161); slightly worse results were obtained in case of the Page model. The highest value of the determination coefficient R^2 (0.996–0.999) and the lowest values of the *RMSE* (0.005–0.0189) during sublimation and convective drying of dragonhead leaves were obtained for the Page model. The Page model very often gave the best match results among the most frequently used empirical models during leaves drying [31,41,42]. The logarithmic model was used to describe the kinetics of mint [43] and rosemary [44] leaves drying.

Table 2. Analysis of models describing the kinetics of freeze-drying of Moldavian dragonhead.

Model	Temperature								
	20 °C			40 °C			60 °C		
	R^2	*RMSE*	χ^2	R^2	*RMSE*	χ^2	R^2	*RMSE*	χ^2
Newton	0.993	0.0234	0.00056	0.987	0.0327	0.00109	0.986	0.0356	0.00130
Page	0.997	0.0067	0.00005	0.999	0.0103	0.00011	0.999	0.0050	0.00003
Henderson and Pabis	0.996	0.0179	0.00033	0.992	0.0252	0.00066	0.992	0.0263	0.00073
Logarithmic	0.998	0.0141	0.00021	0.994	0.0233	0.00058	0.996	0.0201	0.00044
Wang and Singh	0.985	0.0351	0.00128	0.980	0.0404	0.00170	0.994	0.0239	0.00060

Table 3. Statistical analysis of modelsdescribing the kinetics of vacuum drying of Moldavian dragonhead.

Model	Temperature								
	20 °C			40 °C			60 °C		
	R^2	*RMSE*	χ^2	R^2	*RMSE*	χ^2	R^2	*RMSE*	χ^2
Newton	0.996	0.0157	0.00025	0.998	0.0108	0.00012	0.996	0.0180	0.00033
Page	0.997	0.0128	0.00017	0.998	0.0107	0.00012	0.997	0.0157	0.00026
Henderson and Pabis	0.998	0.0111	0.00013	0.999	0.0098	0.00010	0.996	0.0180	0.00034
Logarithmic	0.998	0.0106	0.00012	0.999	0.0098	0.00010	0.997	0.0161	0.00028
Wang and Singh	0.937	0.0641	0.00427	0.932	0.0678	0.00481	0.970	0.0478	0.00242

Table 4. Statistical analysis of modelsdescribingthe kinetics of air drying of Moldavian dragonhead.

Model	Temperature					
	40 °C			60 °C		
	R^2	*RMSE*	χ^2	R^2	*RMSE*	χ^2
Newton	0.976	0.0465	0.0022	0.999	0.0071	0.00005
Page	0.996	0.0189	0.00037	0.999	0.0058	0.00004
Henderson and Pabis	0.982	0.0401	0.00166	0.999	0.0068	0.00005
Logarithmic	0.991	0.0290	0.00089	0.999	0.0033	0.00001
Wang and Singh	0.997	0.0172	0.00031	0.893	0.0857	0.00783

The values of the coefficients occurring in the five analyzed regression equations are presented in Table 5 (sublimation drying), Table 6 (vacuum drying) and Table 7 (convective drying).

Table 5. Values in the models describing the freeze-drying of Moldavian dragonhead.

Temperature	Equation	Coefficient			
		a	***k*(min^{-1})**	***n***	***b***
20 °C	Newton		0.01327		
	Page		0.00643	1.15999	
	Henderson and Pabis	1.05578	0.01398		
	Logarithmic	1.06681	0.01293		−0.02618
	Wang and Singh	−0.00908			0.00002
	Equation	**Coefficient**			
		a	***K***	***n***	***b***
40 °C	Newton		0.01591		
	Page		0.00556	1.24311	
	Henderson and Pabis	1.07812	0.01708		
	Logarithmic	1.08677	0.01608		−0.02042
	Wang and Singh	−0.01071			0.00003
	Equation	**Coefficient**			
		a	***k*(min^{-1})**	***n***	***b***
60 °C	Newton		0.01744		
	Page		0.00596	1.25437	
	Henderson and Pabis	1.08277	0.01881		
	Logarithmic	1.10679	0.01661		−0.04600
	Wang and Singh	−0.01245			0.00004

Table 6. Values in the models describing the vacuum drying of Moldavian dragonhead.

Temperature	Equation	Coefficient			
		a	***k* (min^{-1})**	***n***	***b***
20 °C	Newton		0.01558		
	Page		0.02017	0.94146	
	Henderson and Pabis	0.95767	0.01492		
	Logarithmic	0.96042	0.01452		−0.00774
	Wang and Singh	−0.01009			0.00003
	Equation	**Coefficient**			
		a	***k* (min^{-1})**	***n***	***b***
40 °C	Newton		0.02089		
	Page		0.02193	0.98822	
	Henderson and Pabis	0.98256	0.02053		
	Logarithmic	0.98275	0.02049		−0.00055
	Wang and Singh	−0.01297			0.00004
	Equation	**Coefficient**			
		a	***k* (min^{-1})**	***n***	***b***
60 °C	Newton		0.02265		
	Page		0.01766	1.06203	
	Henderson and Pabis	1.00193	0.02269		
	Logarithmic	1.01051	0.02140		−0.01871
	Wang and Singh	−0.01515			0.00006

Table 7. Values in the models describing the air drying of Moldavian dragonhead.

Temperature	Equation	Coefficient			
		a	$k(\text{min}^{-1})$	*n*	*b*
40 °C	Newton		0.03282		
	Page		0.03659	0.97013	
	Henderson and Pabis	0.99175	0.03255		
	Logarithmic	0.98829	0.03381		0.01052
	Wang and Singh	−0.01895			0.00009
	Equation	**Coefficient**			
		a	$k(\text{min}^{-1})$	*n*	*b*
60 °C	Newton		0.01219		
	Page		0.00277	1.32170	
	Henderson and Pabis	1.08280	0.01312		
	Logarithmic	1.12548	0.01071		−0.07812
	Wang and Singh	−0.00864			0.00002

3.2. Color Assessment

In the whole measuring range (Table 8), dried Moldavian dragonhead leaf material was characterized by a higher brightness *(L*)* and color saturation *(C)* than the raw material. The value of the color shade *(HU)* was lower for the dried material in relation to the raw material. The value of the color coordinate *L** of the dried material decreased with an increase in temperature for sublimation and vacuum drying and increased for the dried material obtained after convective drying. At a given temperature level, the greatest brightness was characteristic of the dried material obtained after sublimation drying, slightly lower values were observed for the dried material obtained as a result of vacuum drying, while the dried material obtained after convective drying had significantly lower values of the *L** color coordinate.

Table 8. Effect of the drying method on the color parameters of leaves from Moldavian dragonhead.

MD*	DT	Dimension of Color			
		L*	C	HU	ΔE
RM		$40.2 \pm 0.43^{d**}$	15.3 ± 0.46^{a}	119.6 ± 1.27^{a}	0
FD	20 °C	58.6 ± 0.57^{c}	23.7 ± 0.59^{b}	112.5 ± 1.02^{e}	20.3 ± 0.50^{ab}
	40 °C	57.8 ± 0.65^{bc}	23.7 ± 0.32^{b}	$110{,}0 \pm 1.59^{f}$	19.7 ± 0.69^{ad}
	60 °C	54.8 ± 0.61^{a}	23.8 ± 0.49^{b}	104.8 ± 1.39^{bc}	17.6 ± 0.42^{e}
VD	20 °C	57.2 ± 0.38^{b}	25.8 ± 0.35^{c}	107.5 ± 1.16^{d}	20.4 ± 0.24^{ab}
	40 °C	54.7 ± 0.67^{a}	26.2 ± 0.34^{c}	$106{,}0 \pm 1.02^{cd}$	18.7 ± 0.34^{cd}
	60 °C	$53{,}0 \pm 0.66^{e}$	27.4 ± 0.57^{d}	102.7 ± 1.07^{b}	18.6 ± 0.34^{c}
AD	40 °C	45.9 ± 0.70^{f}	34.9 ± 0.47^{e}	95.2 ± 0.71^{g}	22.7 ± 0.67^{f}
	60 °C	48.9 ± 0.55^{g}	32.2 ± 0.49^{f}	98.2 ± 1.12^{h}	20.8 ± 0.52^{b}

* MD—Method of drying, DT—Drying temperature, RM—Raw material, FD–Freeze-drying, VD—Vacuum drying, AD—Air drying, *L**—Brightness, *C*—Color saturation, *HU*—Color shade, *ΔE*—Total change in color. ** The values designated by the different small letters (a, b, c, d . . .) are significantly different ($\alpha = 0.05$).

The dried material obtained by sublimation drying at 20 °C and 40 °C was characterized by the highest brightness. The lowest value of this parameter was found in the dried material obtained after convective drying at 40 °C. In this case, the value of the *L** coordinate was lower by about 12.7 units in comparison with the dried material obtained after sublimation drying (20 °C). The saturation of color *(C)* of the dried Moldavian dragonhead leaf material did not depend on the temperature of the sublimation drying. An increase in the temperature of the heating panels contributed to a slight increase in the value of color saturation of the dried material obtained after vacuum drying (these changes were statistically insignificant at 20 °C and 40 °C). Increasing the temperature of the drying air resulted in a decrease in the color saturation during convective drying of the Moldavian dragonhead leaves. The saturation of color closest to the raw material was obtained after sublimation drying (increase by about 8.5 units); the biggest differences occurred after convective drying at 40 °C (increase by about 19.6 units). The dried material from the Moldavian dragonhead, for all the analyzed drying methods, had lower values of color shade (*HU*) than the raw material. As the temperature of the heating panels increased (sublimation and vacuum drying), the value of the color shade decreased. The differences were small at 20 °C and 40 °C and deepened at 60 °C. At each temperature level, the dried material obtained after sublimation drying had higher values of color shade than that obtained by vacuum drying. The values of this color coordinate were lower by about 5 units at 20 °C, 4 units after drying at 40 °C and 2 units at 60 °C. Convective drying resulted in a significant reduction in the color shade values of the dried material, compared to the other two drying methods. Greater changes compared to the raw material were observed at 40 °C. Dried material from the Moldavian dragonhead leaves with the shade of color closest to that of the raw material was obtained after sublimation drying at 20 °C (about 7 units); the greatest difference between the shade of color of the raw material and that of the dried material occurred after convective drying at 40 °C (about 24 units). The total change in the color of the dried material from the Moldavian dragonhead ranged from 17.6 to 22.7. As the temperature increased, regardless of the drying method used, the value of the total change in color (ΔE) decreased. The lowest ΔE values were obtained after sublimation drying at 60 °C, and the highest after convective drying at 40 °C. At a given temperature level, the lowest values of total color change were obtained after sublimation drying, and the highest after convective drying.

The main factors causing adverse changes in the color of the dried leaves are increased temperature and the presence of reactive oxygen in the drying agent. Undesirable changes of color, occurring in dried materials, are mainly due to enzymatic reactions and non-enzymatic browning [45,46]. The color change is also related to the breakdown of pigments, mainly anthocyanins, carotenoids and chlorophylls [47,48]; polyphenolic compounds contained in the raw material; and the breakdown of L-ascorbic acid [49]. The final color of the dried material is also affected by ions of certain metals and the pH of the dried material [50]. Adverse changes in the color of the dragonhead leaves were mainly from the degradation of the chlorophylls contained in them, which contributes to the browning of the dried material [51]. The browning process is accurately reflected by the change in color shade *(HU)* and lightness of the dried material *(L*)*, the value of which decreases as the process temperature increases [52,53]. The chlorophyll content decreases with increasing temperature, process time [54] and the presence of oxygen, which causes the oxidation of unsaturated color compounds contained in the material [55], contributing to unfavorable changes in the color determinants of the dried material.

3.3. Phenolic Profile and Antioxidant Activity

The dried material obtained by the three drying methods studied was characterized by a decrease in total polyphenols (*TPC*), total monomeric anthocyanins (*TAC*) and total flavonoid content (*TFC*) (Table 9). The dried material obtained after sublimation and vacuum drying at 20 °C and 40 °C did not differ significantly in *TPC* content. The total content of polyphenols at 60 °C was lower than at the other two drying temperatures, but the values obtained did not differ significantly in sublimation and vacuum drying. The drying temperature also did not influence the *TPC* content in case of convective drying. However, the *TPC* content obtained after convective drying was significantly lower than that of the other two drying methods (about 9 mg GAE·g DM^{-1} at 40 °C and about 14.5 mg GAE·g DM^{-1} at 60 °C). Sublimation and vacuum drying contributed to about 21%–31% *TPC* losses in dried leaves of Moldavian dragonhead, while convective drying caused losses of about 40%–45% in relation to the raw material.

Table 9. Profileand antioxidant activity of dried leaves from Moldavian dragonhead.

MD *	DT	*TPC* (mg GAE/g DM)	ABTS (EC_{50}; mg DM/mL)	DPPH (EC_{50}; mg DM/mL)	*TAC* (mg Cy-3-G/g DM)	*TFC* (mg QE/g DM)
RM		94.9 ± 4.56^{d}**	9.38 ± 0.378^{c}	17.72 ± 0.327^{d}	6.80 ± 0.230^{c}	307.3 ± 4.34^{c}
FD	20 °C	75.3 ± 4.10^{a}	11.19 ± 0.303^{a}	20.36 ± 0.666^{a}	6.05 ± 0.190^{a}	302.0 ± 3.20^{c}
	40 °C	74.7 ± 2.36^{a}	12.09 ± 0.487^{b}	21.44 ± 0.868^{ab}	5.81 ± 0.191^{a}	292.5 ± 4.39^{b}
	60 °C	67.8 ± 2.50^{b}	13.46 ± 0.213^{d}	21.86 ± 0.297^{b}	4.90 ± 0.216^{b}	273.5 ± 5.24^{d}
VD	20 °C	76.6 ± 1.38^{a}	11.67 ± 0.368^{ab}	20.58 ± 0.801^{a}	5.91 ± 0.293^{a}	288.5 ± 2.41^{ab}
	40 °C	71.8 ± 3.36^{ab}	11.77 ± 0.521^{ab}	21.28 ± 0.719^{ab}	4.97 ± 0.161^{b}	283.7 ± 5.05^{a}
	60 °C	65.2 ± 4.23^{b}	14.93 ± 0.213^{e}	23.38 ± 0.217^{e}	4.24 ± 0.130^{d}	260.8 ± 4.77^{e}
AD	40 °C	56.2 ± 1.08^{c}	15.80 ± 0.391^{f}	24.68 ± 0.466^{c}	3.51 ± 0.123^{e}	200.1 ± 3.71^{f}
	60 °C	51.7 ± 3.55^{c}	17.17 ± 0.443^{g}	24.78 ± 0.559^{c}	2.73 ± 0.082^{f}	182.2 ± 2.27^{g}

* MD—Method of drying, DT—Drying temperature, RM—Raw material, FD–Freeze-drying, VD—Vacuum drying, AD–Air drying, *TPC*—Total phenolics content, ABTS—Antioxidant activity, DPPH—Antioxidant activity, *TAC*—Total anthocyanin content, *TFC*—total flavonoids content. ** The values designated by the different small letters (a, b, c, d ...) are significantly different (α = 0.05).

Increasing the temperature of the sublimation, vacuum and convective drying contributed to a decrease in the total content of anthocyanins in dried dragonhead leaves (except for 20 °C and 40 °C—sublimation drying—where these changes were statistically insignificant). At a given temperature level, the highest *TAC* content was noted for the dried material obtained as a result of sublimation drying, slightly lower after vacuum drying (at 20 °C the changes were statistically insignificant compared to sublimation drying) and the lowest for convective drying. Sublimation drying resulted in a decrease in *TAC* content in the dried dragonhead leaves by about 0.8–1.9 mg Cy-3-G·g DM^{-1}, as compared to the raw material. Vacuum drying resulted in a loss of *TAC* content by about 0.9–2.6 mg Cy-3-G·g DM^{-1}. The dried product obtained after convective drying at 60 °C contained about 2.5 times less *TAC* than the raw material. As the temperature of the sublimation, vacuum and convective drying increased, the total content of flavonoids in the dried dragonhead leaves decreased. These losses were small after sublimation and vacuum drying at 20 °C and 40 °C (about 2%–8%, depending on the temperature and drying method). Increasing the temperature to 60 °C resulted in a decrease in *TFC* content by 24.5 mg QE·g DM^{-1}—sublimation drying—and by 27.7 mg QE·g DM^{-1}—vacuum drying—compared to 20 °C. The dried material obtained by convective drying was characterized by a significant reduction in *TFC* content, in relation to that obtained by the other two methods. The lowest *TFC* content was obtained after convective drying at 60 °C. The loss of this component was then about 40.7% compared to the raw material. In a similar way as described above, the drying method influenced the total content of polyphenols, anthocyanins and flavonoids in dried *Vitexagnus-castus* leaf material [56]. The highest content of these components was found in the dried material obtained after sublimation and vacuum drying. Convective dried material had significantly lower contents of *TPC*, *TAC* and *TFC* [56,57].

The drying of Moldavian dragonhead leaves, regardless of the method used and the drying temperature, resulted in a decrease in the antioxidant capacity of the dried material, compared to the raw material (against ABTS and DPPH radicals). In the whole measuring range, the EC_{50} had a higher value for DPPH radical than for ABTS. As the temperature increases, for all drying methods, the value of EC_{50} of dried material from Moldavian dragonhead leaves increases against ABTS and DPPH (decreasing antioxidant potential). Similarly, the influence of temperature on the DPPH radical scavenging rate in mulberry leaves was described by Ma et al. [58]. The increase in EC_{50} value is small for the dried material obtained at all temperatures used for sublimation drying and vacuum drying at 20 °C and 40 °C, especially for the DPPH radical. The antioxidant potential of the dried material obtained after vacuum drying at 60 °C is slightly higher than that of the convective drying obtained at 40 °C. The lowest antioxidant potential was that of the dried product obtained after convective drying at 60 °C. The value of EC_{50} increased by 1.8 times (ABTS) and 1.4 times (DPPH), respectively. Vuong et al. [56] have shown that sublimation and vacuum drying have little effect on the antioxidant capacity of the dried material; the ABTS and DPPH radical scavenging capacity is significantly reduced in the dried material obtained from convective drying.

3.4. Essential Oils Content

Table 10 lists the 13 main components of the essential oil contained in the raw material and dried leaves of Moldavian dragonhead. The percentage content of these components is from 98.5% to 99.5% of the total amount of obtained essential oil. The main components of the essential oil from Moldavian dragonhead leaves were geranial, geraniol, geranyl acetate, neral and neryl acetate. They belong to the group of non-ring-like monoterpenes (alcohols, ester aldehydes) with a smell similar to lemon and rose. The drying of Moldavian dragonhead leaves resulted in an increase in the content of geraniol, neryl acetate, linalool and, in most cases, geranyl acetate, but a decrease in the content of neral, geranial and β-pinene.

The effect of the method and drying temperature on the content of other compounds present in the oil from the Moldavian dragonhead leaves was ambiguous. As the drying temperature increased, the geranial content decreased. Sublimation and vacuum drying, in the whole temperature range, caused slight losses of geranial content, not exceeding 18%, in relation to its content in the raw material. Convective dried material was characterized by higher losses of this component, which was about 35% at 40 °C and 62% at 60 °C, respectively. An increase in temperature from 20 °C to 60 °C resulted in a 3.4% decrease in neral content for sublimation drying and 4.5% decrease for vacuum drying. An increase in the convective drying temperature from 40 °C to 60 °C reduced the neral content by 5.1%. The geraniol content in the dried material increased with the temperature of sublimation, vacuum and convective drying. At a given temperature level, the highest content of geraniol was found in the dried material obtained after convective drying; the lowest after sublimation drying. The content of geraniol in the dried material obtained after convective drying at 60 °C was about 150% of its content in the raw material. The content of geranyl acetate in the dried material was stable in the range 22%–26% of the total amount of essential oil. The percentage of linalool content in the dried material essential oil increased with the temperature of vacuum and convective drying. In the dried material obtained at 60 °C, it was about seven times higher than in the raw material for vacuum drying, andalmost ten times higher in the case of convective drying. As the drying temperature increased, the content of neryl acetate in the dried material increased, regardless of the drying method used. This increase was the smallest in the dried material obtained after sublimation drying and the largest after convective drying. An increase in geraniol, neryl acetate and geranyl acetate content was also noted by Samadi et al. [59] in the *Dracocephalum kotschyi* Boiss dried material. At 70 °C, they noted significant decreases in the content of these essential oil components. In most cases, the content of essential oil components in the dried leaves was lower in the dried material than in the raw material and decreased as the drying temperature increased [60,61].

Analyzing the changes in the oil content in the dried material from the Moldavian dragonhead leaves, it can be stated that the aldehydes (geranialandneral) are subject to decomposition, leading to the formation of alcohols and esters (geraniol, linalool, neryl acetate and geranial acetate) during the drying process. The total content of essential oils in the dried dragonhead leaves was comparable with their amount in the raw material, after sublimation drying at 20 °C and 40 °C. For the other two drying methods, the total amount of essential oils was lower than in the raw material. As the temperature of vacuum and convective drying increased, the total amount of obtained oils decreased. The lowest content of essential oil was obtained after convective drying at 60 °C; it was 40% lower than in the raw material. In a similar way, the influence of temperature and drying method on the total content of essential oils was described by Arwgyropoulos and Muller [62] for dried material of lemon balm leaves, as well as by Figiel et al. [63] for dried oregano leaves.

Table 10. Oil composition of Moldavian dragonhead leaves influenced by different drying method.

Compound	RI*	RM	Drying Methods							
			FD 20 °C	FD 40 °C	FD 60 °C	VD 20 °C	VD 40 °C	VD 60 °C	AD 40 °C	AD 60 °C
Sabinene	974	$0.5 \pm 0.01^{b**}$	0.5 ± 0.02^{b}	0.4 ± 0.01^{c}	0.3 ± 0.01^{d}	0.6 ± 0.01^{a}	0.6 ± 0.01^{a}	-	0.2 ± 0.03^{e}	0.1 ± 0.02^{f}
β-pinene	980	1.1 ± 0.04^{c}	1.2 ± 0.02^{d}	1.1 ± 0.03^{cd}	1.1 ± 0.02^{c}	0.7 ± 0.03^{a}	0.8 ± 0.04^{b}	0.8 ± 0.03^{b}	0.7 ± 0.03^{a}	0.6 ± 0.05^{a}
Linalool	1102	0.8 ± 0.02^{b}	1.3 ± 0.02^{c}	1.0 ± 0.02^{d}	2.2 ± 0.04^{e}	1.6 ± 0.02^{f}	3.3 ± 0.04^{g}	5.9 ± 0.05^{a}	6.0 ± 0.03^{a}	7.9 ± 0.03^{h}
cis-rose oxide	1108	0.7 ± 0.02^{c}	0.9 ± 0.01^{d}	0.6 ± 0.01^{a}	0.7 ± 0.01^{c}	0.7 ± 0.03^{c}	0.5 ± 0.02^{ab}	0.5 ± 0.03^{b}	0.4 ± 0.01^{d}	-
cis-limonene oxide	1150	0.7 ± 0.02^{c}	0.5 ± 0.02^{b}	0.4 ± 0.01^{a}	0.5 ± 0.01^{b}	0.5 ± 0.03^{b}	0.5 ± 0.02^{b}	-	0.5 ± 0.03^{b}	0.4 ± 0.03^{ab}
cis-chrysanthenol	1162	1.1 ± 0.04^{a}	1.3 ± 0.03^{b}	1.1 ± 0.03^{a}	1.1 ± 0.03^{a}	1.2 ± 0.03^{b}	1.5 ± 0.03^{c}	1.0 ± 0.01^{a}	0.9 ± 0.02^{c}	0.7 ± 0.01^{c}
trans-limonene oxide	1168	1.3 ± 0.02^{c}	1.6 ± 0.03^{d}	1.6 ± 0.03^{d}	1.3 ± 0.02^{c}	1.0 ± 0.02^{b}	0.8 ± 0.01^{a}	1.0 ± 0.02^{b}	0.7 ± 0.04^{e}	0.8 ± 0.02^{a}
Citronellal	1178	0.6 ± 0.01^{c}	0.4 ± 0.02^{a}	-	-	0.4 ± 0.02^{a}	0.4 ± 0.01^{a}	-	0.1 ± 0.02^{b}	-
Neral	1238	18.6 ± 0.11^{b}	17.5 ± 0.2^{c}	15.9 ± 0.21^{d}	14.1 ± 0.06^{a}	16.3 ± 0.21^{e}	13.9 ± 0.17^{a}	11.8 ± 0.10^{f}	14.5 ± 0.19^{g}	9.4 ± 0.15^{h}
Geraniol	1260	20.7 ± 0.18^{c}	22.2 ± 0.13^{d}	23.5 ± 0.07^{a}	22.9 ± 0.10^{e}	23.2 ± 0.12^{a}	24.0 ± 0.08^{b}	24.2 ± 0.10^{b}	26.8 ± 0.17^{f}	30.5 ± 0.20^{g}
Geranial	1276	27.3 ± 0.14^{a}	26.3 ± 0.19^{b}	23.2 ± 0.15^{c}	24.8 ± 0.13^{d}	25.4 ± 0.21^{e}	25.9 ± 0.18^{f}	22.5 ± 0.15^{g}	17.7 ± 0.12^{h}	10.4 ± 0.13^{i}
Neryl acetate	1360	2.1 ± 0.02^{a}	3.2 ± 0.02^{b}	5.1 ± 0.01^{c}	5.9 ± 0.02^{d}	3.9 ± 0.02^{e}	5.3 ± 0.01^{f}	6.6 ± 0.02^{g}	9.1 ± 0.02^{h}	11.3 ± 0.01^{i}
Geranyl acetate	1378	23.2 ± 0.18^{b}	22.5 ± 0.12^{c}	24.9 ± 0.12^{a}	24.3 ± 0.2^{d}	23.6 ± 0.23^{e}	22.1 ± 0.22^{f}	24.8 ± 0.04^{a}	21.2 ± 0.09^{g}	26.2 ± 0.15^{h}
Total		98.8	99.4	98.7	99.0	99.1	99.5	99.2	98.9	98.5
Essential oil (%)		0.62 ± 0.015^{a}	0.62 ± 0.018^{a}	0.63 ± 0.020^{a}	0.57 ± 0.0213^{b}	0.52 ± 0.018^{c}	0.49 ± 0.009^{d}	0.46 ± 0.018^{e}	0.42 ± 0.023^{f}	0.37 ± 0.016^{g}

* RI—Retention index, RM—Raw material, FD—Freeze-drying, VD—Vacuum drying, AD—Air drying. ** The values designated by the different small letters (a, b, c, d …) in the lines of the table are significantly different ($\alpha = 0.05$).

4. Conclusions

The best match of the analyzed models describing the change in reduced water content as a function of drying time was obtained using a logarithmic model in case of vacuum drying, and a Page model for sublimation and vacuum drying. At a given temperature level, sublimation and vacuum drying were characterized by comparable process times. As the temperature increased, the determinants of the color of dried dragonhead leaves, the content of the phenolic compounds, the antioxidant capacity and the composition of essential oils differed to a greater extent from the values of these parameters for the raw material before drying. The differences were small for the dried material obtained after sublimation and vacuum drying, especially at 20 °C and 40 °C, and quite significant after convective drying. If the main emphasis is placed on the quality characteristics of the dried material, and the drying time plays a smaller role, it is recommended to carry out the process of drying Moldavian dragonhead leaves by sublimation or vacuum at a heating-panel temperature not exceeding 40 °C. If the drying time plays a major role, the drying heating process should be carried out by convection at 60 °C.

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Abbreviations

χ^2	chi-squared test
ABTS	antioxidant activity (ability to neutralize free radicals against ABTS)
C	color saturation
DM	dry mass
DPPH	antioxidant activity (ability to neutralize free radicals against DPPH)
HU	color shade
MR	reduced water content
RMSE	root mean square error
TAC	total monomeric anthocyanins
TFC	total flavonoids content
TPC	total phenolics content
ΔE	total change in color

References

1. Dastmalchi, K.; Dorman, H.J.D.; Laakso, I.; Hiltunen, R. Chemical composition and antioxidative activity of Moldavian balm (*DracocephalummoldavicaL.*) extracts. *Food Sci. Technol. – LEB.* **2007**, *40*, 1655–1663. [CrossRef]
2. Popova, O.I.; Nikitina, A.S.; Markova, O.M. Studies of iridoids from *Dracocephalum moldavica* cultivated in the Stavropol region. *Pharm. Chem. J.* **2008**, *42*, 39–42. [CrossRef]
3. Omidbaigi, R.; Yavari, S.; Hassani, M.E.; Yavari, S. Induction of autotetraploidy in dragonhead (*Dracocephalum moldavica*) by colchicine treatment. *J. Fruit Ornam. Plant Res.* **2010**, *18*, 23–35.
4. Sultan, A.; Bahang, A.; Aisa, H.A.; Eshbakova, K.A. Flavonoids from *Dracocephalummoldavica*. *Chem. Nat. Compd.* **2008**, *44*, 336–337. [CrossRef]
5. Dziki, D.; Miś, A.; Gładyszewska, B.; Laskowski, J.; Kwiatkowski, S.; Gawlik-Dziki, U. Physicochemical and grinding characteristics of dragonhead seeds. *Int. Agrophys.* **2013**, *27*, 403–408. [CrossRef]
6. Janmohammadi, M.; Yousefzadeh, S.; Sabaghnia, N. Chemical comparison of essential oils in dragonhead (*Dracocephalum moldavica* L.) samples grown in different areas. *J. Essent. Oil-Bear Plant* **2018**, *21*, 950–962. [CrossRef]

7. Dmitruk, M.; Weryszko-Chmielewska, E. Morphological differentiationand distribution of non-glandular and glandular trichomes on *Dracocephalum moldavicum* L. shoots. *ActaAgrobotanica* **2010**, *63*, 97–110.
8. Yousefzadeh, S.; Modarres-Sanavy, S.A.M.; Sefidkon, F.; Asgarzadeh, A.; Ghalavand, A.; Sadat-Asilan, K. Effects of Azocompostandurea on the herbage yield and contents and compositions of essentialoils from two genotypes of dragonhead (*Dracocephalum moldavica* L.) in two regions of Iran. *Food Chem.* **2013**, *138*, 1407–1413. [CrossRef]
9. Yang, L.N.; Xing, J.G.; He, C.H.; Wu, T. The phenolic compounds from *Dracocephalum moldavica* L. *Biochem. Syst. Ecol.* **2014**, *54*, 19–22. [CrossRef]
10. Dziki, D.; Cacak-Pietrzak, G.; Gawlik-Dziki, U.; Sułek, A.; Kocira, S.; Biernacka, B. Effect of moldavian dragonhead (*Dracocephalum moldavica* L.) leaves on the baking properties of wheat flour and quality of bread. *CyTA-J Food* **2019**, *17*, 536–543. [CrossRef]
11. Wójtowicz, A.; Oniszczuk, A.; Oniszczuk, T.; Kocira, S.; Wojtunik, K.; Mitrus, M.; Kocira, A.; Widelski, J.; Skalicka-Woźniak, K. Application of Moldavian dragonhead (*Dracocephalum moldavica* L.) leaves addition as a functional component of nutritionally valuable corn snacks. *J. Food Sci. Tech. Mys.* **2017**, *54*, 3218–3229. [CrossRef] [PubMed]
12. Shofian, N.; Hamid, A.; Osman, A.; Saari, N.; Anwar, F.; Dek, M.; Hairuddin, M. Effect of freeze-drying on the antioxidant compounds and antioxidant activity of selected tropical fruits. *Int. J. Mol. Sci.* **2011**, *12*, 4678–4692. [CrossRef] [PubMed]
13. Oliveira, S.; Brandão, T.; Silva, C. Influence of drying processes and pretreatments on nutritional and bioactive characteristics of dried vegetables: A review. *Food Eng. Rev.* **2016**, *8*, 134–163. [CrossRef]
14. Kumar, C.; Karim, M.; Joardder, M. Intermittent drying of food products: A critical review. *J. Food Eng.* **2014**, *121*, 48–57. [CrossRef]
15. Karam, M.; Petit, J.; Zimmer, D.; Djantou, D.; Scher, J. Effects of drying and grinding in production of fruit and vegetable powders: A review. *J. Food Eng.* **2016**, *188*, 32–49. [CrossRef]
16. Bozin, B.; Mimica-Dukic, N.; Samojlik, I.; Jovin, E. Antimicrobial and antioxidant properties of rosemary and sage (*Rosmarinus officinalis L.* and *Salvia officinalis L., Lamiaceae*) essential oils. *J. Agric. Food Chem.* **2007**, *55*, 7879–7885. [CrossRef]
17. Gutierrez, J.; Barry-Ryan, C.; Bourke, P. The antimicrobial efficacy of plant essential oil combinations and interactions with food ingredients. *Int. J. Food Microbiol.* **2008**, *124*, 91–97. [CrossRef]
18. Pirbalouti, A.; Rahimmalek, M.; Malekpoor, F.; Karimi, A. Variation in antibacterial activity, thymol and carvacrol contents of wild populations of *Thymus daenensis subsp. Daenensis Celak*. *Plant Omics J.* **2011**, *4*, 209–214.
19. Kubra, R.; Mohan Rao, J. An impression on current developments in the technology, chemistry, and biological activities of ginger (*Zingiberofficinale Roscoe*). *Crit. Rev. Food Sci. Nutr.* **2012**, *52*, 651–688. [CrossRef]
20. Rahimmalek, M.; Goli, S. Evaluation of six drying treatments with respect to essential oil yield, composition and color characteristics of *Thymysdaenensis subsp. daenensis*. Celak leaves. *Ind. Crops Prod.* **2013**, *42*, 613–619. [CrossRef]
21. Guo, C.; Zhang, N.; Liu, C.; Xue, J.; Chu, J.; Yao, X. Qualities and antioxidant activities of lotus leaf affected by different drying methods. *Acta Physiol. Plant.* **2020**, *42*. [CrossRef]
22. Yap, J.Y.; Hii, C.L.; Ong, S.P.; Lim, K.H.; Abas, F.; Pin, K.Y. Effects of drying on total polyphenols content and antioxidant properties of carica papaya leaves. *J. Sci. Food Agric.* **2020**, *100*, 2932–2937. [CrossRef] [PubMed]
23. Choo, C.O.; Chua, B.L.; Figiel, A.; Jałoszyński, K.; Wojdyło, A.; Szumny, A.; Łyczko, J.; Chong, C.H. Hybrid drying of murrayakoenigii leaves: Energy consumption, antioxidant capacity, profiling of volatile compounds and quality studies. *Processes* **2020**, *8*, 240. [CrossRef]
24. do NascimentoSilveiraDorneles, L.; Luís Duarte Goneli, A.; Andrea Lima Cardoso, C.; Bezerra da Silva, C.; RosemariHauth, M.; Cardoso Oba, G.; Schoeninger, V. Effect of air temperature and velocity on drying kinetics and essential oil composition of piper umbellatum L. leaves. *Ind. Crop. Prod.* **2019**, *142*. [CrossRef]
25. de Souza, R.A.; Melo, E.C.; de Ávila, M.B.R.; Gonzaga, D.A.; Sperotto, N.C.Z.; Carneiro, A.P.S. Intermittent drying of clove basil leaves: Process optimization and essential oil yield. *RevistaBrasileira De Engenharia Agricola e Ambiental* **2020**, *24*, 209–215. [CrossRef]
26. Rudy, S. Energy consumption in the freeze - and convection-drying of garlic. *TEKA Comm. Mot. Power Ind. Agric.* **2009**, *9*, 259–266.

27. Ertekin, C.; Yaldiz, O. Drying of eggplant and selection of a suitable thin layer drying model. *J. Food Eng.* **2004**, *63*, 349–359. [CrossRef]
28. Rasouli, M.; Seiiedlou, S.; Ghasemzadeh, H.; Nalbandi, H. Convective drying of garlic (*Allium sativum L.*): Part I: Drying kinetics, mathematical modeling and change in color. *Aust. J. Crop Sci.* **2011**, *13*, 1707–1714.
29. Rayaguru, K.; Routray, W. Mathematical modeling of thin layer drying kinetics of stone apple slices. *Int. Food Res. J.* **2012**, *19*, 1503–1510.
30. Demir, V.; Gunhan, T.; Yagcioglu, A.K.; Degirmencioglu, A. Mathematical modeling and the determination of some quality parameters of air-dried bay leaves. *Biosyst. Eng.* **2004**, *88*, 325–335. [CrossRef]
31. Sarimeseli, A. Microwave drying characteristics of coriander (*Coriandru msativum L.*) leaves. *Energy Convers. Manag.* **2011**, *52*, 1449–1453. [CrossRef]
32. Henderson, S.M.; Pabis, S. Grain drying theory. II. Temperature effects on drying coefficients. *J. Agric. Eng. Res.* **1961**, *6*, 169–174.
33. Dandamrongrak, R.; Young, G.; Mason, R. Evaluation of various pre-treatments for the dehydration of banana and selection of suitable drying models. *J. Food Eng.* **2002**, *95*, 139–146. [CrossRef]
34. Wang, C.Y.; Singh, R.P. Use of variable equilibrium moisture content in modeling rice drying. *Trans. ASAE* **1978**, *11*, 668–672.
35. Mohammadi, A.; Rafiee, S.; Emam-Djomeh, Z.; Keyhani, A. Kinetic models for colour changes in kiwifruit slices during hot air drying. *World J. Agric. Sci.* **2008**, *84*, 376–383.
36. Singleton, V.L.; Rossi, J.A. Colorimetry of total phenolics with phosphomolybdic-phosphotungstic acid reagents. *Am. J. Enol. Viticult.* **1965**, *16*, 144–158.
37. Re, R.; Pellegrini, N.; Proteggente, A.; Pannala, A.; Yang, M.; Rice-Evans, C. Antioxidant activity applying an improved ABTS radical cationdecolorization assay. *Free Rad. Biol. Med.* **1999**, *26*, 1231–1237. [CrossRef]
38. Brand-Williams, W.; Cuvelier, M.E.; Berset, C. Use of a free radical method to evaluate antioxidant activity. *LWT - Food Sci. Technol.* **1995**, *28*, 25–30. [CrossRef]
39. Bahorun, T.; Luximon-Ramma, A.; Crozier, A.; Aruoma, O.I. Total phenol, flavonoid, proanthocyanidin and vitamin C levels and antioxidant activities of Mauritian vegetables. *J. Sci. Food Agric.* **2004**, *84*, 1553–1561. [CrossRef]
40. Giusti, M.M.; Wrolstad, R.E. Characterization and measurement of anthocyanins by UV–visible spectroscopy. In *Current Protocols in Food Analytical Chemistry*, 2nd ed.; Wrolstad, R.E., Ed.; John Wiley & Sons: New York, NY, USA, 2002; units I1.1.1–I1.1.8.
41. Alibas, I. Energy consumption and colour characteristics of nettle leaves during microwave, vacuum and convective drying. *Biosyst. Eng.* **2007**, *96*, 49–52. [CrossRef]
42. Costa, A.B.S.; Freire, F.B.; Ferreira, M.C.; Freire, J.T. Convective drying of regular mint leaves: Analysis based on fitting empirical correlations, response surface methodology and neural networks. *Acta Sci.-Technol.* **2014**, *36*, 271–278. [CrossRef]
43. Hussein, I.; Mamman, M.; Abdulrasheed, M. Effect of varying drying temperature on the antibacterial activity of Moringaoleifera Leaf (Lam). *IOSR J. Pharm. Biol. Sci.* **2015**, *10*, 39–43.
44. Arslan, D.; Ozcan, M. Evaluation of drying methods with respect to drying kinetics, mineral content and colour characteristics of rosemary leaves. *Energy Covers. Manag.* **2008**, *49*, 1258–1264. [CrossRef]
45. Vadivambal, R.; Jayas, D. Changes in quality of microwave treated agricultural products-a review. *Biosyst. Eng.* **2007**, *98*, 1–16. [CrossRef]
46. Cecchini, M.; Contini, M.; Massantini, R.; Monarca, D.; Moscetti, R. Effects of controlled atmospheres and low temperature on storability of chestnuts manually and mechanically harvested. *Postharvest Biol. Technol.* **2011**, *61*, 131–136. [CrossRef]
47. Roshanak, S.; Rahimmalek, M.; Goli, S. Evaluation of seven different drying treatments in respect to total flavonoid, phenolic, vitamin C content, chlorophyll, antioxidant activity and color of green tea (*Camellia sinensis or C. assamica*) leaves. *Food Sci. Technol.* **2015**, *53*, 721–729. [CrossRef] [PubMed]
48. Madhava, N.M.; Vedashree, M.; Satapathy, P.; Khanum, H.; Ramsamy, R.; Hebbar, H. Effect of drying methods on the quality characteristics of dill (*Anethumgraveolens*) greens. *Food Chem.* **2016**, *192*, 849–856. [CrossRef] [PubMed]
49. Lozano, J.; Ibarz, A. Colour changes in concentrated fruit pulp during heating at high temperatures. *J. Food Eng.* **1997**, *31*, 365–373. [CrossRef]

50. García-Viguera, C.; Zafrilla, P.; Romero, F.; Abellán, P.; Artés, F.; Tomás-Barberán, F.A. Colour stability of strawberry jamas affected by cultivar and storage temperature. *J. Food Sci.* **1999**, *64*, 243–247. [CrossRef]
51. Di Cesare, L.F.; Forni, E.; Viscardi, D.; Nani, R.C. Changes in the chemical composition of basil caused by different drying procedures. *J. Agric. Food Chem.* **2003**, *51*, 3575–3581. [CrossRef]
52. Razak, N.A.; Shaari, A.R.; Jolkili, M.; Leng, L.Y. Drying curves and colour changes of Cassia Alata leaves at different temperatures. In *Proceedings of the MATEC Web of Conferences, Phuket, Thailand, 1–2 May 2016*; EDP Science: Paris, Fance, 2016; Volume 78, p. 1020. [CrossRef]
53. Telfser, A.; Galindo, F.G. Effect of reversible permeabilization in combination with different drying methods on the structure and sensorial quality of dried basil (*Ocimum basilicum L.*) leaves. *LWT - Food Sci. Technol.* **2019**, *99*, 148–155. [CrossRef]
54. Lin, X.; Zhang, L.; Lei, H.; Zhang, H.; Cheng, Y.; Zhu, R.; Ruan, R. Effect of drying technologies on quality of green tea. *Int. Agric. Eng. J.* **2010**, *19*, 30–37.
55. Negi, P.S.; Roy, S.K. Effect of drying conditions on quality of green leaves during long term storage. *Food Res. Int.* **2001**, *34*, 283–287. [CrossRef]
56. Vuong, Q.V.; Zammit, N.; Munro, B.R.; Murchie, S.; Bowyer, M.C.; Scarlett, C.J. Effect of drying conditions on physicochemical and antioxidant properties of *Vitex Agnus-Castus* leaves. *J. Food Process. Preserv.* **2015**, *39*, 2562–2571. [CrossRef]
57. Rabeta, M.; Lai, S. Effects of drying, fermented and unfermented tea of *Ocimum tenuiflorum Linn.* on the antioxidant capacity. *Int. Food Res. J.* **2013**, *20*, 1601–1608.
58. Ma, Q.; Santhanam, R.K.; Xue, Z.; Guo, Q.; Gao, X.; Chen, H. Effect of different dryingmethods on the physicochemical properties and antioxidant activities of mulberry leaves polysaccharides. *Int. J. Biol. Macromol.* **2018**, *119*, 1137–1143. [CrossRef]
59. Samadi, L.; Larijani, K.; Badi, H.N.; Mehrafarin, A. Qualitative and quantitative variations of the essential oils of *Dracocephalu mkotschyi Boiss.*as affected by different drying methods. *J. Food Preserv.* **2018**, *42*, e13816. [CrossRef]
60. Szumny, A.; Figiel, A.; Gutiérrez-Ortíz, A.; Carbonell-Barrachina, A. Composition of rosemary essential oil (*Rosmarinus officinalis*) as affected by drying method. *J. Food Eng.* **2010**, *97*, 253–260. [CrossRef]
61. Calín-Sánchez, A.; Lech, K.; Szumny, A.; Figiel, A.; Carbonell-Barrachina, A. Volatile composition of sweet basil essential oil (*Ocimum basilicum L.*) as affectedby drying method. *Food Res. Int.* **2012**, *48*, 217–225. [CrossRef]
62. Argyropoulos, D.; Müller, J. Changes of essential oil content and composition during convective drying of lemon balm. *Ind. Crops Prod.* **2014**, *52*, 118–124. [CrossRef]
63. Figiel, A.; Szumny, A.; Gutiérrez-Ortíz, A.; Carbonell-Barrachina, A. Composition of oregano essential oil (*Origanum vulgare*) as affected by drying method. *J. Food Eng.* **2010**, *98*, 240–247. [CrossRef]