Hybrid Drying of Murraya koenigii Leaves: Energy Consumption, Antioxidant Capacity, Profiling of Volatile Compounds and Quality Studies

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Keywords: GC-MS, antioxidant capacity, energy consumption, murraya koenigii, CPD-MVFD, MVD, hybrid drying

Abstract:

This study aims to reduce the amount of specific energy consumed during the drying of fresh Murraya koenigii leaves by comparing four drying methods: (1) convective hot-air drying (CD; 40, 50 and 60 °C); (2) single-stage microwave-vacuum drying (MVD; 6, 9 and 12 W/g); (3) two-stage convective hot-air pre-drying followed by microwave-vacuum finishing?drying (CPD-MVFD; 50 °C, 9 W/g); and (4) freeze-drying as a control in the analysis sections. The drying kinetics were also modelled using thin-layer models. The quality parameters of dried M. koenigii leaves were measured including total polyphenolic content (TPC), antioxidant capacity (ABTS and FRAP), profiling of volatile compounds, colour analysis and water activity analysis. Results showed that CPD-MVFD effectively reduced the specific energy consumption of CD at 50 °C by 67.3% in terms of kilojoules per gram of fresh weight and 48.9% in terms of kilojoules per gram of water. The modified Page model demonstrated excellent fitting to the empirical data obtained. FD showed promising antioxidant activity. The major contributor of antioxidant capacity was TPC. The volatile compounds profiled by gas chromatography-mass spectrometry, namely, ?-phellandrene (31%), ?-pinene (19.9%), and sabinene (16%) were identified as the major compounds of dried M. koenigii leaves. Colour analysis showed MVD's high performance in preserving the colour parameters of M. koenigii leaves under all conditions. The colour parameters were correlated to the antioxidant capacity and TPC. Water activity analysis showed that the water activity of M. koenigii leaves for all drying methods indicating that the conditions were microbiologically and shelf-stable. Pearson correlation showed the colour parameters of the leaves had a strong correlation to TPC. Overall, MVD showed promising energy consumption reduction and recovery in TPC and volatile compounds.

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Hybrid Drying of *Murraya koenigii* Leaves: Energy Consumption, Antioxidant Capacity, Profiling of Volatile Compounds and Quality Studies

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Abstract: This study aims to reduce the amount of specific energy consumed during the drying of fresh *Murraya koenigii* leaves by comparing four drying methods: (1) convective hot-air drying (CD; 40, 50 and 60 °C); (2) single-stage microwave-vacuum drying (MVD; 6, 9 and 12 W/g); (3) two-stage convective hot-air pre-drying followed by microwave-vacuum finishing–drying (CPD-MVFD; 50 °C, 9 W/g); and (4) freeze-drying as a control in the analysis sections. The drying kinetics were also modelled using thin-layer models. The quality parameters of dried *M. koenigii* leaves were measured including total polyphenolic content (TPC), antioxidant capacity (ABTS and FRAP), profiling of volatile compounds, colour analysis and water activity analysis. Results showed that CPD-MVFD effectively reduced the specific energy consumption of CD at 50 °C by 67.3% in terms of kilojoules per gram of fresh weight and 48.9% in terms of kilojoules per gram of water. The modified Page model demonstrated excellent fitting to the empirical data obtained. FD showed promising antioxidant activity. The major contributor of antioxidant capacity was TPC. The volatile compounds profiled by gas chromatography-mass spectrometry, namely, β-phellandrene (31%), α-pinene (19.9%), and sabinene (16%) were identified as the major compounds of dried *M. koenigii* leaves. Colour analysis showed MVD's high performance in preserving the colour parameters of *M. koenigii* leaves under all conditions. The colour parameters were correlated to the antioxidant capacity and TPC. Water activity analysis showed that the water activity of *M. koenigii* leaves for all drying methods indicating that the conditions were microbiologically and shelf-stable. Pearson correlation showed the colour parameters of the leaves had a strong correlation to TPC. Overall, MVD showed promising energy consumption reduction and recovery in TPC and volatile compounds.

Keywords: hybrid drying; MVD; CPD-MVFD; murraya koenigii; energy consumption; GC-MS; antioxidant capacity

1. Introduction

Oxidative stress is a condition of oxidative damage when the critical balance between free radical generation and antioxidant defenses is disrupted [1]. Short-term oxidative damage may cause human health conditions, such as trauma, infection and heat injury [2]. Hence, antioxidants play an important role in stabilising free radicals by donating an electron to neutralise them. Plants contain a promising amount of antioxidants to protect and preserve their physical and metabolic integrity [3]. *Murraya koenigii*, which is commonly known as curry leaf, is commonly used as a natural flavouring agent because of its aromatic scent and medicinal value [4,5]. *M. koenigii* belongs to the Rutaceae family, which is native in Malaysia, India and other South Asian countries [6]. *M. koenigii* leaves contain several bioactive compounds that possess anti-tumour, antioxidant, anti-inflammatory and hypoglycemic properties [7]. Essential oils, which can be found in the leaves of *M. koenigii*, are significant contributors to antioxidant activity [8–10]. However, the proper preservation of these pharmacological properties and their corresponding bioactive compounds through the processing of herbal plants remains challenging.

Drying is a challenging process to retain pharmacological properties. It is usually performed to preserve herbal plants and their bioactive compounds in post-harvesting. This process involves the removal of water from herbal plants to reduce their microbiological activity and preserve their bioactive compounds [11,12]. Traditional drying methods, such as sun drying and air drying, have been used for a long time for the dehydration of food due to their simplicity and low operating cost. However, open-air drying and long drying duration cause food to be contaminated with dust and microbes [13]. Another type of drying method that is widely used in dehydrating agricultural products is convective hot-air drying (CD). This drying method involves two types of moisture diffusions during heat transfer. The first type is external diffusion, where the surface moisture content diffuses to the drying medium; the second type is internal diffusion, where internal moisture content diffuses out to the drying surface [14]. One of the disadvantages of CD is long drying duration due to the internal diffusion process. The drying process requires higher energy compared with other production processes due to the extremely low energy efficiency of dryers and high latent heat of water evaporation [15]. Drying consumes 10% to 15% of the total national industrial energy demand in the USA and 20% to 25% in Europe [16]. Thus, the drying industry is faced with challenges to resolve this problem by optimising drying conditions or developing an alternative drying method, such as hybrid drying.

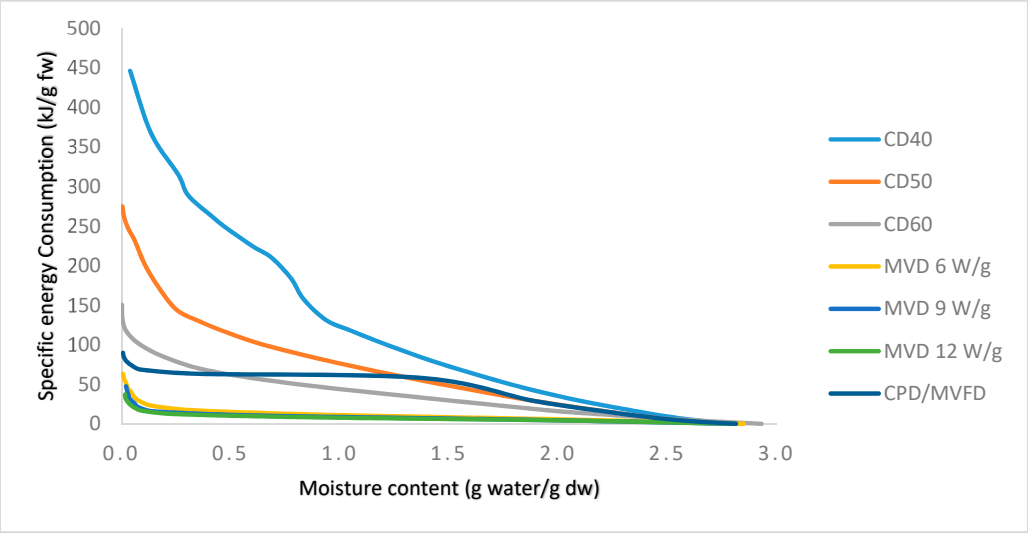
Hybrid drying combines two or more drying methods into one drying system. Hybrid drying systems occur in two types, namely, single-stage hybrid drying and two-stage hybrid drying. Single-stage hybrid drying combines two drying methods, such as microwave drying and vacuum drying, into a one-unit system called microwave vacuum drying (MVD). However, one of the disadvantages of MVD is that the intensive water evaporation from the leaves may exceed the capacity of the vacuum pump. Thus, this technique requires the reduction of raw materials or an increase in the capacity of the vacuum pump [17]. This problem can be overcome using CD as a pre-drying to efficiently remove excess moisture and decrease the load of the vacuum system in MVD. Convective hot-air pre-drying before MVD (CPD-MVFD) allows a reduction in the total cost of the drying process and improves the quality of products [18]. To overcome the energy consumption issue of the drying process, two-stage hybrid drying methods, such as MVD and CPD-MVFD, were used in this study to compare its specific energy consumption with commercial drying method (CD) and reduce energy consumption during the drying process.

In this present study, four drying methods, namely, freeze-drying (FD), CD, MVD, and two-stage hybrid CPD-MVFD were used to estimate and reduce energy consumption during the drying of *M. koenigii* leaves without compromising the quality of the dried leaves. Five quality parameters, including antioxidant capacity (ABTS and FRAP), total polyphenolic content (TPC) analysis, profiling of volatile compounds, colour analysis and water analysis, were analysed. The Pearson correlation analysis was performed to study the correlation between ABTS, FRAP, TPC, volatile compounds and colour parameters and to identify the main contributor to the antioxidant capacity.

2. Results and Discussion

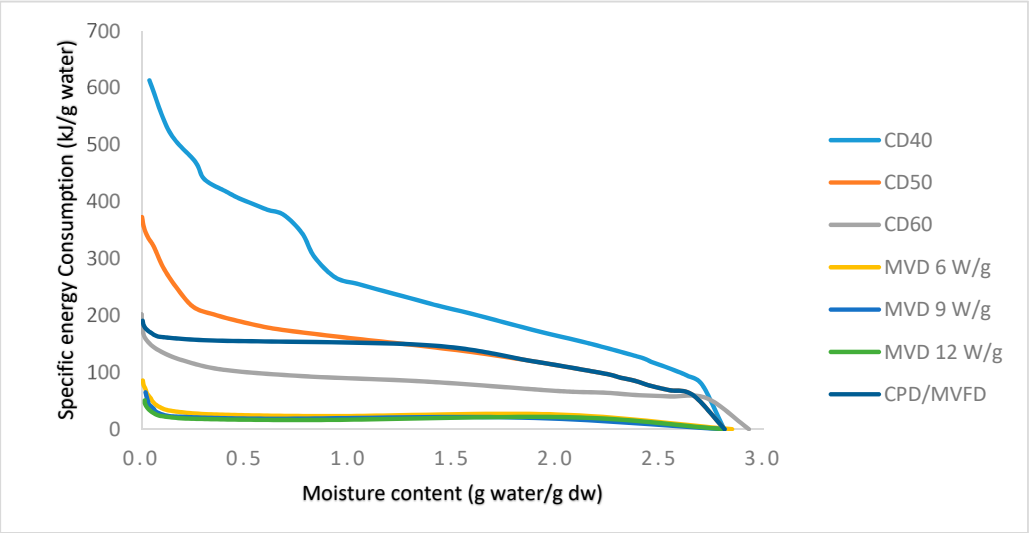
2.1. Energy Consumption of Convective Hot-Air Drying, Microwave Vacuum Drying and Convective Hot-Air Pre-Drying followed by Microwave Vacuum Finishing-Drying

The energy expressed in terms of kilojoules per gram of fresh weight (fw) and kilojoules per gram of water are presented in Figure 1a,b. The initial specific energy consumption was 0 kJ/g fw for Figure 1a and 0 kJ/g water for Figure 1b at the initial moisture content of 3.0 g water/g dw and increased exponentially as the moisture content reached approximately 0.01 g water/g dw. The results showed that all temperatures for CD exhibited the highest energy consumption compared with that for MVD and CPD-MVFD. CD at 40 °C had the highest energy consumption with a final energy consumption of 492.11 kJ/g fw and 676.27 kJ/g water. On the other hand, MVD at 12 W/g consumed the least energy with an energy consumption of 25.98 kJ/g fw and 35.43 kJ/g water. With MVFD at 9 W/g combined with CPD at 50 °C, the energy consumption was successfully reduced from 274.86 kJ/g fw and 372.92 kJ/g water to 84.11 kJ/g fw and 161.60 kJ/g water, respectively. This reduction resulted from the fact that after reaching a moisture content of 1.4 g water/g dw, convective method was replaced by microwave-vacuum method, which was more energy-efficient. The course of specific energy consumption during MVFD applied after CPD has been highlighted by the dark blue line in Figure 1. Moreover, drying time plays a vital role in energy consumption. The shorter the drying time, the lower the energy consumption even though increased power is required. Increased drying intensity is a factor that reduced drying time. This factor is associated with increased moisture diffusion, which is enhanced by the increased temperature of the dried material. Moisture contained in the sample can be removed rapidly by increasing the drying temperature or microwave power, hence reducing drying time. This condition also increases the dried material temperature. The same phenomenon occurred on garlic and pomegranate [17,19]. The final specific energy consumption in terms of a gram of fw and gram of water is presented in Table 1.



(**a**)

Figure 1. *Cont.*



(**b**)

Figure 1. Specific energy consumption of *M. koenigii* (**a**) in kJ/g fw and in (**b**) kJ/g water for convective hot air drying (CD), microwave vacuum drying (MVD) and microwave vacuum finishing-drying (MVFD) applied after CPD during CPD-MVFD.

Table 1. Final specific energy consumption of dried *M. koenigii* leaves subjected to CD, MVD, and CPD-MVFD.

Drying Methods	Final Energy Consumption, E_m (kJ/g fw)	Final Energy Consumption, E_w (kJ/g water)
CD40	492.11	676.27
CD50	274.86	372.92
CD60	130.28	174.78
MVD 6 W/g	63.15	85.49
MVD 9 W/g	39.01	53.25
MVD 12 W/g	25.98	35.43
CPD-MVFD	84.11	161.60

MVD consumed the lowest energy compared with CD and CPD-MVFD. However, the maintenance and installation cost of MVD for an industrial application are considerably high due to the vacuum pump and the microwave system. Thus, CD can be introduced as a pre-drying step to aid in removing excess moisture from the surface of the material; finally, drying can be finished with MVD to remove internal moisture content by utilising microwave energy [17,20]. In this research, the combined hybrid drying method had reduced the total drying time of CD at 50 °C from 570 min to 138 min (Table 2) and thus reduced the specific energy consumption by 67.3% (kJ/g fw) and 48.9% (kJ/g water). This occurrence was due to the penetration of microwave energy into the inner layer of the leaves, forcing the moisture out to the surface of the leaves through evaporation and diffusion [21].

2.2. Modelling of Drying Kinetics for Dried M. koenigii Leaves

Table 2 shows the statistical parameters, drying time and model constants describing the drying kinetics of *M. koenigii* leaves under thin-layer models (Lewis Newton, Midilli–Kucuk and Modified Page model). The highest value of the coefficient of determination (R^2) and the lowest values for both root–mean–square error (RMSE) and chi-square coefficient (χ^2) demonstrate good fitting of the model to the empirical data [22]. The values of R^2, RMSE and χ^2 obtained in this study showed that the modified Page model had best fitting to the empirical data. Parameter a represents the initial moisture ratio (M_R), and the values were one for the CD and MVD. However, for CPD-MVFD, a varied

(0.3834) after convective hot-air pre-drying. The model constants b, k and n represent the drying rate. The greater the values are, the faster the drying process is, as proven in this study. As the drying rate of the MVD increased from 0.0646 g water/g dw to 0.1551 g water/g dw, k and n increased from 0.0400 to 0.0876 and 1.5961 to 1.8593, respectively. The same trend was also observed in the CD.

Table 2. Drying time, statistical parameters and model constants describing the drying kinetics of *Muraya koenigii* leaves.

Drying Method	Models	R^2	RMSE	χ^2	a	b	k	n	t
CD40	Lewis	0.9955	0.0315	0.0267	-	-	0.0035	-	1020
	Midilli–Kucuk	0.8694	0.3776	3.8489	0.2729	-0.0001	0.0001	0.9827	
	Modified Page	0.9976	0.0159	0.0068	1.0000	-	0.0082	0.8505	
CD50	Lewis	0.9968	0.0221	0.0107	-	-	0.0070	-	570
	Midilli–Kucuk	0.9872	0.0411	0.0371	0.9222	-0.0055	-0.0014	1.0709	
	Modified Page	0.9969	0.0219	0.0105	1.0000	-	0.0064	1.0197	
CD60	Lewis	0.9969	0.0280	0.0118	-	-	0.0146	-	270
	Midilli–Kucuk	0.9901	0.0374	0.0210	0.9400	-0.0106	-0.0015	1.1841	
	Modified Page	0.9977	0.0192	0.0055	1.0000	-	0.0090	1.1179	
MVD 6 W/g	Lewis	0.9807	0.0473	0.0268	-	-	0.1409	-	44
	Midilli–Kucuk	0.9993	0.0082	0.0008	1.0018	0.0003	0.0390	1.6135	
	Modified Page	0.9994	0.0108	0.0014	1.0000	-	0.0400	1.5961	
MVD 9 W/g	Lewis	0.9751	0.0547	0.0299	-	-	0.2229	-	27
	Midilli–Kucuk	0.9996	0.0070	0.0005	1.0007	0.0006	0.0611	1.7918	
	Modified Page	0.9996	0.0113	0.0013	1.0000	-	0.0625	1.7681	
MVD 12 W/g	Lewis	0.9683	0.0664	0.0441	-	-	0.2878	-	18
	Midilli–Kucuk	0.9996	0.0069	0.0005	1.0015	0.0007	0.0867	1.8782	
	Modified Page	0.9996	0.0102	0.0010	1.0000	-	0.0876	1.8593	
CPD50-MVFD9W/g	Lewis	0.9686	0.2332	0.3807	-	-	0.6985	-	138
	Midilli–Kucuk	0.9996	0.0027	0.0001	0.3835	0.0002	0.3697	1.0608	
	Modified Page	0.9660	0.2331	0.3802	0.3834		1.0349	0.6605	

R^2—determination coefficient, RMSE – root–mean–square error, χ^2—chi-square coefficient, a, b, k, n—model constants, t—total drying time (min), CD—convective hot-air drying, MVD–MVD, CPD–convective hot-air pre-drying, MVFD—microwave vacuum finishing-drying

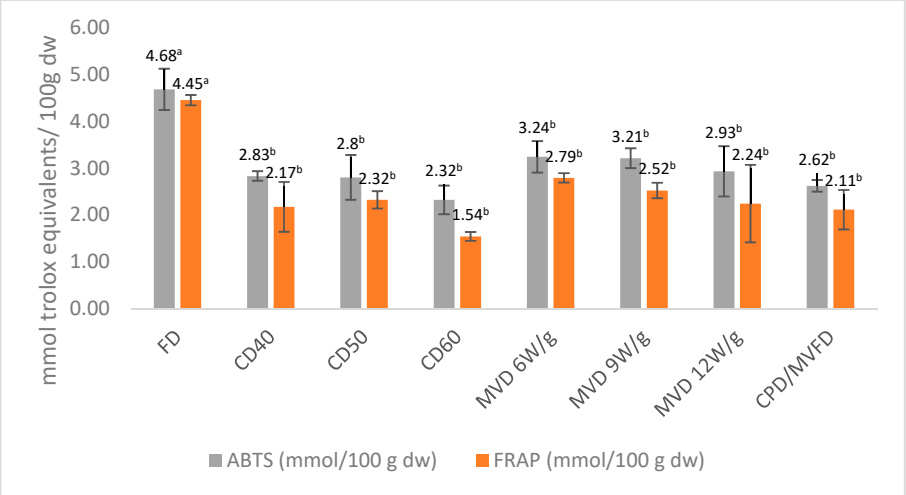
2.3. Antioxidant Capacity and Total Polyphenolic Content (TPC)

TPC and antioxidant capacity were analysed to ensure the quality of the dried leaves. Figure 2a shows the effect of FD, CD, MVD, and CPD-MVFD on the antioxidant capacity of *M. koenigii* leaves. The highest antioxidant capacity was achieved for FD with 4.68 ± 0.44 and 4.45 ± 0.11 mmol/100 g dw for 2,2′-azinobis(3-ethylbenzthiazoline-6-sulfonic acid) (ABTS) and ferric reducing antioxidant power (FRAP), respectively. FD acted as a control in this study as it is a non-thermal drying method that removes water content from a solid phase to the gas phase through sublimation. Thus, FD has the highest antioxidant capacity as FD is effective in preserving heat-sensitive antioxidant compounds [14].

The loss of antioxidant capacity was found in the samples dried with thermal drying methods, where the highest loss was found in CD under all conditions and the least loss was found in MVD under all conditions. This result might be caused by thermal antioxidant degradation and oxidative degradation [20]. The intensity of drying methods, such as drying temperature and microwave power was found to influence the antioxidant capacity as the drying temperature increased from 40 °C to 60 °C, the antioxidant capacity reduced from 2.83 ± 0.10 mmol/100 g dw to 2.32 ± 0.31 mmol/100 g dw for ABTS and from 2.17 ± 0.53 mmol/100 g dw to 1.54 ± 0.10 mmol/100 g dw for FRAP. The same phenomenon was found in MVD, an increase in microwave power from 6 W/g to 12 W/g decreased the antioxidant capacity by 9% and 19% for ABTS and FRAP, respectively. This finding proves that excess heat during the drying process causes the thermal degradation of antioxidant capacity [18].

Figure 2b shows the TPC of dried *M. koenigi* leaves. The highest retention of TPC was FD (1267.76 mg/100 g dw), followed by MVD at 6 W/g (875.37 mg/100 g dw). The lowest TPC was found in CD at 50 °C with a TPC concentration of 600.49 mg/100 g dw. The lowest retained TPC could be due to the long exposure time to thermal treatment, causing the oxidative degradation of polyphenolic

compounds [20]. Increasing drying intensity could decrease the antioxidant capacity and TPC of *M. koenigii* leaves. This occurrence might be due to the prolonged exposure to a high temperature, causing the degradation of heat-sensitive antioxidant compounds [23].



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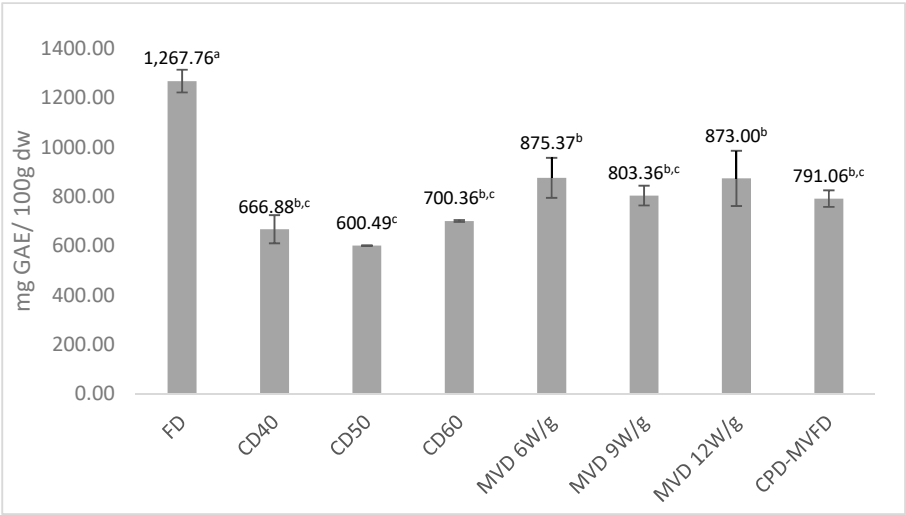

(**b**)

Figure 2. Effect of the FD, CD, MVD, and CPD-MVFD on the antioxidant capacity (**a**) and total polyphenolic content (**b**) of dried *M. koenigii* leaves. (**a**): The same letters next to the bars indicate no significant differences in mean values at the level of 5%, according to Tukey's significant difference test; (**b**): The same letters next to the bars indicate no significant differences in mean values at the level of 5%, according to Tukey's significant difference test.

2.4. Analysis of Volatile Compounds of Different Drying Methods through Gas Chromatography-Mass Spectrometry (GC-MS)

Table 3 shows the retention time, Kovats index in experimental and literature values and quantity for each of the volatile compounds. Kovats index was used in this study to define the retention of compounds analysed through GC-MS by bracketing them with the nearest straight alkane hydrocarbons chain. Twenty-six volatile compounds were identified in fresh *M. koenigii* leaves in this study, with a total quantity of 8.599 mg/g dw, as listed in Table 3. The identified volatile compounds primarily consisted of monoterpenoids and their oxygenated derivatives. The top three major volatile compounds identified and quantified in *M. koenigii* were β-phellandrene (31%; 2.647 mg/g dw), α-pinene (19.9%;

1.711 mg/g dw) and sabinene (16%; 1.374 mg/g dw). These compounds possess a notable effect as antioxidants, which might be contributing to the antioxidant activity in this study [24,25].

A notable loss of volatile content was observed in all the dried samples. In MVD, microwave power with 12 W/g retained the highest volatile content amongst the others. Increasing the intensity of microwave power from 6 W/g to 12 W/g in the MVD increased volatile content from 4.982 mg/g dw to 5.330 mg/g dw. This promising trend occurred because the increase in the intensity of the microwave power reduced the total drying time. Hence, the exposure time for high degradative temperature was reduced [20]. In CD, a decreasing trend of volatile content was observed when the hot-air temperature increased from 40 °C (4.687 mg/g dw) to 60 °C (3.420 mg/g dw). This result might be due to the loss of individual volatile compounds when the drying temperature increased. A similar trend was also found in the drying of *Cassia alata* [20].

In this study, CPD-MVFD was expected to retain high volatile contents as it shortened thermal treatment, which assured that the volatile contents were not exposed to high drying temperature for a prolonged drying period. However, the volatile contents for dried *M. koenigii* leaves using CPD-MVFD were lost by 66% in comparison to fresh curry leaves. The total concentration of volatile contents decreased from 8.599 mg/g dw to 2.882 mg/g dw after the drying treatment was applied. Further to this, Chua et al. [18] obtained the highest retention of volatile compounds for the drying treatment of *Phyla nodiflora* using CPD at 50 °C followed by the MVFD at 6 W/g. Hence, the morphology of the plant material may be one of the factors affecting the retention of volatile compounds. Overall, MVD at 12 W/g had the promising quantification of volatile compounds of 5.33 mg/g dw.

Table 3. Volatile compounds of dried *M. koenigii* leaves dried using different methods at different conditions and identified and quantified through GC-MS.

No.	Compound	Retention Time	Kovat Index		Quantity (mg/g dw)								
			Exp.	Lit.	Fresh	FD	CD40	CD50	CD60	MVD 6 W/g	MVD 9 W/g	MVVD 12 W/g	CPD-MVFD
1	α-Thujene	7.70	927	929	0.098	0.071	0.098	0.059	0.079	0.107	0.051	0.108	0.058
2	α-Pinene	7.92	933	937	1.711	0.903	0.816	0.453	0.604	0.966	0.467	1.006	0.509
3	Sabinene	9.23	973	975	1.374	0.509	0.242	0.117	0.163	0.320	0.167	0.372	0.128
4	β-Pinene	9.33	976	979	0.355	0.197	0.167	0.100	0.121	0.205	0.098	0.214	0.098
5	β-Myrcene	9.84	991	991	0.197	0.073	0.067	0.039	0.056	0.077	0.041	0.080	0.041
6	α-Phellandrene	10.28	1004	1002	0.330	0.148	0.128	0.080	0.097	0.168	0.084	0.164	0.077
7	α-Terpinene	10.72	1016	1017	0.163	0.095	0.142	0.092	0.104	0.147	0.073	0.143	0.075
8	*p*-Cymene	10.99	1024	1025	0.015	0.014	0.049	0.025	0.034	0.036	0.017	0.033	0.022
9	β-Phellandrene	11.15	1028	1029	2.647	1.269	1.184	0.720	0.867	1.437	0.752	1.413	0.682
10	β-Ocimene	11.86	1048	1050	0.331	0.141	0.110	0.063	0.074	0.123	0.070	0.145	0.060
11	γ-Terpinene	12.23	1058	1060	0.240	0.136	0.213	0.139	0.157	0.221	0.116	0.216	0.111
12	*trans*-Sabinene hydrate	12.55	1067	1070	0.087	0.094	0.114	0.071	0.080	0.055	0.072	0.073	0.082
13	Terpinolene	13.31	1088	1088	0.064	0.032	0.044	0.029	0.033	0.044	0.027	0.047	0.022
14	Linalool	13.70	1099	1099	0.112	0.137	0.153	0.103	0.119	0.077	0.113	0.105	0.120
15	*trans*-*p*-Menth-2-en-1-ol	14.52	1140	1140	0.035	0.053	0.047	0.032	0.034	0.028	0.035	0.038	0.037
16	Terpinen-4-ol	16.54	1178	1177	0.133	0.181	0.181	0.125	0.132	0.108	0.129	0.137	0.131
17	α-Terpineol	17.03	1192	1190	0.045	0.062	0.055	0.035	0.043	0.033	0.043	0.038	0.043
18	Bornyl acetate	20.34	1287	1284	0.042	0.038	0.040	0.020	0.015	0.034	0.032	0.037	0.020
19	β-Ionone	22.67	1363		0.046	0.028	0.027	0.026	0.028	0.020	0.027	0.027	0.027
20	Caryophyllene	24.32	1426	1419	0.323	0.293	0.320	0.204	0.231	0.329	0.294	0.403	0.196
21	Humulene	25.06	1460	1454	0.087	0.077	0.090	0.054	0.065	0.090	0.080	0.104	0.052
22	β-Eudesmene	25.70	1492	1489	0.031	0.045	0.049	0.030	0.037	0.051	0.052	0.066	0.033
23	Valencene	25.87	1500	1494	0.031	0.146	0.122	0.076	0.090	0.134	0.137	0.167	0.086
24	Spathulenol	27.27	1586	1578	0.007	0.035	0.047	0.021	0.029	0.032	0.038	0.033	0.031
25	Caryophyllene oxide	27.37	1591	1582	0.015	0.040	0.058	0.027	0.036	0.035	0.042	0.038	0.036
26	Selin-6-en-4α-ol	28.41	1624	1636	0.080	0.168	0.124	0.071	0.091	0.106	0.140	0.121	0.107
	Total				8.599 [a]	4.986 [b]	4.687 [b]	2.809 [c]	3.420 [c]	4.982 [b]	3.198 [c]	5.330 [b]	2.882 [c]

Exp.—experimental, Lit.—literature, FD—freeze drying, CD—convective hot-air drying, MVD—microwave vacuum drying, CPD—convective hot-air pre-drying, MVFD—microwave vacuum finishing drying; mean values followed by the same letters were not significantly different at the level of 5%, according to Tukey's significant difference test.

2.5. Colour Analysis of M. koenigii Leaves

The colour of *M. koenigii* leaves was measured to study the correlation among the colour parameters of dried *M. koenigii* leaves, antioxidant activity and TPC. The colour parameters of *M. koenigii* leaves after each drying treatment are shown in Table 4. L^* describes the lightness of the dried leaves; the higher the value is, the brighter the dried leaves are. a^* describes the greenness or redness of the dried leaves; the lower the value is, the greener the dried leaves are. b^* indicates the blueness of the dried leaves and yellowness when the value is increasing [18]. In this study, freeze-dried leaves recorded the highest L^* value (lightest), whereas CD at 50 °C yielded the lowest L^* value (darkest). An increase in the drying intensity of CD from 50 °C to 60 °C increased L^* from 42.37 ± 0.188 to 43.67 ± 0.202. The drying duration was shortened due to the increased intensity, which reduced the exposure to oxidation reaction [26]. The same phenomena occurred in MVD. When the microwave power was increased from 9 W/g to 12 W/g, the brightness increased from 44.79 ± 0.133 to 45.66 ± 0.036.

Table 4. Colour parameters of *M. koenigii* leaves after the drying treatment.

Drying Conditions	Colour Parameters		
	L^*	a^*	b^*
FD	46.43 ± 0.097 ^{a}	−7.84 ± 0.142 ^{a}	14.66 ± 0.216 ^{b,c}
CD40	43.42 ± 0.198 ^{d}	−4.01 ± 0.115 ^{e}	13.22 ± 0.458 ^{d}
CD50	42.37 ± 0.188 ^{e}	−3.23 ± 0.177 ^{f}	13.00 ± 0.458 ^{d}
CD60	43.67 ± 0.202 ^{d}	−3.91 ± 0.139 ^{e}	14.21 ± 0.391 ^{c}
MVD 6 W/g	45.46 ± 0.139 ^{b}	−6.63 ± 0.136 ^{b}	15.15 ± 0.129 ^{a,b}
MVD 9 W/g	44.79 ± 0.133 ^{c}	−6.17 ± 0.092 ^{c}	14.62 ± 0.344 ^{b,c}
MVD 12 W/g	45.66 ± 0.036 ^{b}	−6.49 ± 0.178 ^{b}	15.30 ± 0.057 ^{a}
CPD-MVFD	44.77 ± 0.121 ^{c}	−4.46 ± 0.502 ^{d}	14.72 ± 0.252 ^{a,b,c}

CD—convective hot-air drying, MVD—microwave vacuum frying, CPD—convective hot-air pre-drying, MVFD—microwave vacuum finishing drying. All values are expressed in mean ± standard deviation; mean value with the same letters, within the same column, were not significantly different at the level of 5%, according to Tukey's significant difference test. L^*—lightness and darkness; a^*—greenness and redness; b^*—yellowness and blueness.

The loss of greenness is related to the degradation of chlorophylls to pheophytins. Pheophytins represent the magnesium-free derivatives of chlorophylls; during its degradation, chlorophyll is assumed to undergo acid-catalysed transformation into pheophytin due to cellular acids [26]. In this process, the magnesium ion of chlorophylls tends to displace with two hydrogens, leading to the formation of pheophytins. The loss of magnesium ions causes the green colour to convert into olive-brown [27]. A factor causing chlorophyll degradation is thought to be associated with the increment of drying temperature and microwave power. This idea was confirmed in the CD and MVD results in this study. As the drying intensity was elevated from 40 °C to 60 °C and from 6 W/g to 12 W/g, a^* increased from −4.01 ± 0.115 to −3.91 ± 0.139 and from −6.63 ± 0.136 to −6.49 ± 0.178, respectively. The lowest a^* was found in FD, as freeze drying is a heatless drying method preventing chlorophyll degradation.

The yellowness or blueness of the curry leaves can be identified using parameter b^*. Carotenoid is one of the compounds causing the leaves to appear yellow. Carotenoid degradation decreases yellowness [26]. Therefore, MVD at 12 W/g could preserve carotenoids well as it had the highest b^* value of 15.30 ± 0.057. CD in all studied conditions had the highest reduction of b^* values, where the colour of leaves changed to slightly blue.

2.6. Water Activity of M. koenigii Leaves

The water activity results of each drying condition are shown in Table 5. The lowest water activity was obtained from MVD at 12 W/g, and the highest was found in CD at 40 °C which were 0.3863 ± 0.001 and 0.4451 ± 0.007, respectively. Increasing the drying temperature of CD from 40 °C to 60 °C improved microbial growth control. The same trend was found in MVD when an increase in microwave power

from 6 W/g to 12 W/g decreased the water activity from 0.3937 ± 0.005 to 0.3863 ± 0.001. Furthermore, the water activity of the dried product was improved using CPD-MVFD instead of CD at 50 °C as evidenced by reducing its value from 0.4173 ± 0.005 to 0.4142 ± 0.001. This improvement could be due to the permeability of microwaves, which effectively removed internal moisture to maintain a low water activity. In summary, the water activity below 0.6 obtained under all conditions of FD, CD, MVD, and CPD-MVFD might imply the microbiological stability [28]. However, further affirmation should be conducted in the future work through microbial test.

Table 5. Water activity of dried *M. koenigii* leaves at different drying conditions.

Drying Conditions	Water Activity, a_w
FD	0.4071 ± 0.001 ^{b,c,d}
CD40	0.4451 ± 0.007 ^{a}
CD50	0.4173 ± 0.005 ^{b}
CD60	0.4291 ± 0.013 ^{a,b}
MVD 6 W/g	0.3937 ± 0.005 ^{c,d}
MVD 9 W/g	0.3894 ± 0.006 ^{d}
MVD 12 W/g	0.3863 ± 0.001 ^{d}
CPD-MVFD	0.4142 ± 0.001 ^{b,c}

CD—convective hot-air drying, MVD—microwave vacuum drying, CPD—convective hot-air pre-drying, MVFD—microwave vacuum finishing drying. All values are expressed in mean ± standard deviation; mean value with the same letters, within the same column, were not significantly different at the level of 5%, according to Tukey's significant difference test

2.7. Correlation Analysis between ABTS, FRAP, TPC, Volatile Compounds, and ColourParameters

The Pearson correlation analysis was performed to determine the correlation between ABTS, FRAP, TPC, volatile compounds and colour parameters. The Pearson coefficients (*r*) for each of the pairs are shown in Table 6. Based on the result, TPC was highly correlated to ABTS and FRAP assays with the Pearson coefficients (*r*) of 0.900 and 0.886, respectively. Besides, the colour parameters of the leaves had a strong correlation to TPC, which proved that the colour parameters of the leaves might affect the yield of TPC.

Table 6. Pearson correlation analysis.

	ABTS	FRAP	TPC	L^*	Volatile	a^*	b^*
ABTS	1						
FRAP	0.992 **	1					
TPC	0.900 **	0.886 **	1				
L^*	0.671	0.643	0.888 **	1			
Volatile	0.484	0.445	0.553	0.627	1		
a^*	−0.798 *	−0.758 *	−0.891 **	−0.939 **	−0.658	1	
b^*	0.265	0.236	0.583	0.868 **	0.423	−0.757 *	1

** Correlation is significant at the $p < 0.01$ (2-tailed test); * correlation is significant at the level of $p < 0.05$ (2-tailed test); TPC—Total polyphenolic content; FRAP—ferric reducing antioxidant power; ABTS—2,2′-azinobis(3-ethylbenzthiazoline-6-sulfonic acid); L^*—lightness and darkness; a^*—greenness and redness; b^*—yellowness and blueness.

3. Material and Methods

3.1. Chemical Reagents

Methanol, hydrochloric acid, Folin–Ciocalteu reagent, sodium carbonate, 6-hydroxyl-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox), 2,2′ -azinobis(3-ethylbenzthiazoline-6-sulfonic acid) (ABTS), potassium persulphate, ferric reducing antioxidant power (FRAP), 2,4,6-tripyridyl-1,3,5-triazine (TPTZ), iron (III) chloride, gallic acid and sodium

sulphate were purchased from Sigma-Aldrich (Steinheim, Germany). Cyclohexane and 2-undecanone were provided by United Quantum Factory (Wrocław, Poland)

3.2. Sample Preparation

Approximately 5 kg of fresh *M. koenigii* leaves were purchased from Ukay Nursery (Kuala Lumpur, Malaysia) and identified at the Forest Research Institute Malaysia (FRIM) with the voucher number of 031/18. The bone drying weight of *M. koenigii* leaves were obtained using an oven dryer at 105 °C for 24 h, in accordance with the ASTM standard (D1348-94) [15].

3.3. Drying of Fresh M. koenigii Leaves

Fresh *M. koenigii* leaves were dried using the four drying methods, namely, CD, VMD, CPD-MVFD, and FD. Freeze-dried samples were used as the control samples [17,19,29].

3.3.1. Convective Hot-Air Drying (CD)

Approximately 40 g of fresh *M. koenigii* leaves were dried using a convective hot-air dryer designed and constructed in the Institute of Agriculture Engineering (Wrocław, Poland) at 40, 50 and 60 °C. The leaves were placed on a wire mesh tray and spread evenly. The wire mesh tray was placed on top of the drying chambers. Weight loss was determined at 5 min interval for the first 1 h, at 30 min interval for the next 5 h and an hour interval until the mass loss difference was 0.05 g or less using an analytical balance (PS600/C/2, Radwag , Wrocław, Poland).

3.3.2. Microwave Vacuum Drying (MVD)

Approximately 40 g of fresh *M. koenigii* leaves were dried using a vacuum microwave dryer (Plazmatronika, Wrocław, Poland) with 6, 9 and 12 W/g microwave power. The leaves were first placed in an organic glass container connected to a vacuum system and then rotated at a constant speed of 6 rpm throughout the drying process to prevent the local overheating of leaves. The temperature of the leaves was measured immediately after the samples were taken out of the dryer. The weight loss of the leaves was recorded for every 4, 3, and 2 min intervals for 6, 9 and 12 W/g, respectively, until the difference in the mass loss was 0.05 g or less using an analytical balance (PS600/C/2, Radwag, Wrocław, Poland).

3.3.3. Convective Hot-Air Pre-Drying and Microwave Vacuum Finishing-Drying (CPD-MVFD)

M. koenigii leaves were first subjected to CPD by using a convective hot-air dryer at 50 °C. The partially dried samples were then subjected to a vacuum microwave dryer (Plazmatronika, Wrocław, Poland) at 9 W/g to thoroughly dry the leaves. A convective hot-air temperature of 50 °C and a microwave wattage of 9 W/g were used in CPD-MVFD to ensure the good quality of the dried product [18,30].

3.3.4. Freeze Drying (FD)

M. koenigii leaves were dehydrated using a freeze dryer (OE-950, Hungary) at a vacuum pressure of 65 Pa. The freezing temperature was −60 °C. The heating plate was set to 30 °C for sublimation.

3.4. Energy Consumption and Specific Energy Consumption of CD, MVD, and CPD-MVFD

Energy consumption (E) is the energy used expressed in kilojoules. Specific energy consumption is an energy performance indicator to evaluate or measure the performance of energy efficiency. In this study, the specific energy consumption of dried *M. koenigii* leaves was expressed in kilojoule per gram of fw (E_m) and kilojoule per gram of water (E_w). The energy consumption and specific energy consumption of CD, MVD, and CPD-MVFD at all drying conditions were determined using the equations shown in Sections 3.4.1–3.4.3 [17,18].

3.4.1. Energy Consumption in Convective Hot-Air Drying Method

Energy consumed (E_{CD}, during CD (kJ)) was calculated using Equation (1):

$$E_{CD} = \left(\frac{N_f}{6} + N_h\right) \times t \tag{1}$$

where N_f is the power consumption by fans blowing air to six pipes (kW), N_h is the power consumption of the electric heater (kW) and t is the drying time (s).

3.4.2. Energy Consumption in Microwave Vacuum Method

The energy consumed during MVD (kJ) was calculated using Equation (2).

$$E_{MVD} = \left(\frac{N_M}{\eta_M} + N_v + N_e\right) \times t \tag{2}$$

where N_M is the output power (kW), and η_M is the efficiency of magnetrons. N_v is the power consumption by the vacuum pump (kW), N_e is the power consumption by the electric engine rotating the container (kW), and t is the time (s).

3.4.3. Specific energy Consumption

Specific energy consumption was expressed in (a) the ratio of energy consumption to the initial mass, $E_m\left(\frac{kJ}{g\ fw}\right)$, and (b) the ratio of energy consumption to the mass of water removed from the material, E_w $(\frac{kJ}{g\ water})$. Equations (3) and (4) show the specific energy consumption for CD, and Equations (5) and (6) show the specific energy consumption for MVD.

$$E_{m_{CD}} = \frac{E_{CD}}{m} \tag{3}$$

$$E_{w_{CD}} = \frac{E_{CD}}{W} \tag{4}$$

$$E_{m_{MVD}} = \frac{E_{MVD}}{m} \tag{5}$$

$$E_{w_{MVD}} = \frac{E_{MVD}}{W} \tag{6}$$

3.5. Modelling of Drying Kinetics

The drying kinetics was modelled in this study to understand the transport mechanism, simulate or scale up the entire optimisation process and control the operating conditions [31]. Therefore, the drying kinetics of *M. koenigii* leaves dried with CD, MVD, and CPD-MVFD at all drying conditions was determined based on the mass loss throughout the drying process. Drying curves were plotted as a function of moisture ratio (M_R) against time. M_R was determined using Equation (7).

$$M_R = \frac{M_i - M_e}{M_o - M_e} \tag{7}$$

where M_i is the moisture content at the respective time (g water/g dw), M_e is the equilibrium moisture content (g water/g dw) representing the lowest moisture content obtainable at equilibrium under the drying conditions used and M_o is the initial moisture content (g water/g dw)

The drying rate (D_R) was calculated using Equation (8):

$$D_R = \frac{M_t - M_{t+\Delta t}}{\Delta t} \tag{8}$$

where M_t is the moisture content at the respective time (t_i), $M_{t+\Delta t}$ is the moisture content at respective time + 1 (t_{i+1}) and Δt is the time difference.

Lewis Newton (Equation (9)), Modified Page (Equation (10)) and Midilli–Kucuk (Equation (11)) were adopted in this study to describe the drying kinetics of dried *M. koenigii* leaves. These three thin-layer models have been frequently used to model the drying kinetics of leaves. They have shown good fitting in describing the drying kinetics of leaves and have been used to simulate or scale up the entire drying process for further optimisation [32–34].

$$M_R = \exp(-kt) \tag{9}$$

$$M_R = a\exp\left((-kt)^n\right) \tag{10}$$

$$M_R = a\exp(-kt^n) + bt \tag{11}$$

where M_R denotes the moisture ratio, a, b denotes the model constants, k denotes the drying constant, n is the dimensionless empirical constant and t is the drying time.

Goodness of fitting was evaluated and compared using the statistical measures, such as R^2, RMSE and χ^2. Goodness of fit was identified based on the highest R^2 value and the lowest RMSE and χ^2.

3.6. Total Polyphenolic Content (TPC) and Antioxidant Capacity Analysis of Dried M. koenigii Leaves

3.6.1. Extraction of TPC from M. koenigii Leaves

The extraction of TPC from dried leaves of *M. koenigii* was conducted using the procedure described by Chua et al. [23] with some modifications. Approximately 0.5 g of powdered sample was extracted with 9 mL of 80% methanol acidified with 1% hydrochloric acid. The extraction was performed in an ultrasonic bath (Sonic 6D; Polsonic, Warsaw, Poland) for 15 min with a frequency of 50 Hz under room temperature. The extract solution was stored at 4 °C overnight and sonicated again at the same extraction conditions. The extracts were centrifuged at 10,000 rpm for 5 min (MPW-350R; Warsaw, Poland) and subjected to TPC and antioxidant capacity analysis.

3.6.2. TPC Analysis

TPC was determined using the Folin–Ciocalteu method described by Hamrouni et al. [35] and Wojdyło et al. [22] with some modifications. Approximately 0.1 mL of sample extract was mixed with 2 mL of distilled water and 0.2 mL of Folin–Ciocalteu reagent. The mixture was then incubated for 3 min at room temperature and added to 1 mL of 20% sodium carbonate. TPC was determined after 1 h of incubation at room temperature in the dark. Absorbance values were determined using UV-2401 PC spectrophotometer (Shimadzu, Kyoto, Japan) at 765 nm. The results were generated in triplicates and expressed in gallic acid equivalence per 100 g of dry weight (mg GAE/100 g dw).

3.6.3. Antioxidant Capacity Analysis

The in vivo mechanisms of action of the antioxidant-protecting effect of *M. koenigii* leaves are complex. Therefore, at least two or more *in vitro* antioxidant assays must be conducted to evaluate different aspects of the reactivity of compound (s) toward reactive oxygen species and reactive nitrogen species [36,37]. Two antioxidant assays, namely, ABTS radical scavenging method and FRAP, were conducted in this study to quantify the antioxidant capacity of *M. koenigii* leaves.

ABTS Radical Scavenging Assay

ABTS radical scavenging assay was determined following the procedure described by Chua et al. [20]. ABTS was dissolved in distilled water at a final concentration of 7 mM and then mixed with 2.45 mM potassium persulphate to obtain ABTS radical cation (ABTS). The mixture was incubated in the dark at room temperature for 12–16 h. Thereafter, the mixture was diluted with

distilled water until an absorbance value of 0.700 ± 0.02 at 734 nm was reached using a UV-2401 PC spectrophotometer (Shimadzu, Kyoto, Japan). Subsequently, 3 mL of the diluted radical solution was added to 30 µL of the extracted supernatant. Absorbance values were determined at 734 nm after 6 min. All determined values were expressed in µM Trolox per 100 g of dry weight and measured in duplicate.

FRAP Assay

FRAP assay was performed following the procedure described by Benzie et al. [38]. FRAP reagent was prepared by mixing acetate buffer (300 µM at a pH 3.6) with 10 µM of TPTZ solution, which was subsequently added with 40 µM of HCl and 20 µM of $FeCl_3$ at a ratio of 10:1:1 (v/v/v). Approximately 3 mL of FRAP reagent was then added to 1 mL of the sample solution and mixed well. Absorbance values were read at 593 nm after 10 min. A standard curve was plotted using different Trolox concentrations. All readings were expressed in µM Trolox per 100 g of dry weight, and the results were obtained in duplicate.

3.7. Hydrodistillation of Volatile Compounds/Essential Oils

The essential oil was extracted using the hydrodistillation method via Deryng type apparatus, as described by Wróblewska et al. [39]. Approximately 1 g of *M. koenigii* leaves was transferred into a 150 mL round bottom flask with 50 mL of distilled water. The bottom flask was connected to the apparatus and heated using a heating mantle. Distillation was finished after 30 min. The essential oils were collected from the three-way tap. Approximately 1 mL of a mixture of cyclohexane and 2-undecanone (concentration 5 mg/mL) (UQF, Wrocław, Poland) was added into the samples as an internal standard for the identification and quantification of volatile compounds using GC-MS. The collected essential oils were dried using anhydrous Na_2SO_4 and stored in the fridge for the profiling of volatile compounds.

Identification and Quantification of Volatile Compounds Using GC-MS

The detailed analysis of the chemical composition of *M. koenigii* leaves was carried out using a gas chromatograph coupled to a mass spectrometer (Shimadzu 2020 Kyoto Japan). The separation was performed on ZB-5 (Phenomenex, Torrance, CA, USA) fused silica capillary tubes column (30 m × 0.25 mm ID × 0.25 µm film thickness). The stationary phase consisted of 5% phenyl and 95% dimethylpolysiloxane. The mass spectrometer was equipped with a quadrupole analyser set at 1508 for all analyses at an electron multiplier voltage of 1350 V. Scanning (1 scan/s) was performed in the range of 39–450 *m*/*z* by using electron impact (*EI*) ionisation at 70 eV. The components were identified by comparison of: (a) the obtained *EI* mass spectra with NIST17 (NIST MS Search 2.0d software, Gaithersburg, MD, USA); (b) the retention times of the authentic standards and analysed signals; (c) the retention Kovats indices (RI exp.) and theoretical value (RI lit. from Adams [40], MassFinder and NIST17 entries). For the calculation of retention indices, homologous series of C7–C20 *n*-alkanes (UQF Wrocław, Poland) were used. The carrier gas was helium with a flow rate of 1 mL/min. The starting temperature of the isolation was 50 °C (2 min), which was raised to 130 °C at 4 °C/min and then reached a final temperature of 270 °C in 10 °C/min. This temperature was retained for 5 min. Injector and detector temperatures were 220 and 300 °C, respectively. The volume of the injected solution was 1 µL with a split ratio 1:40. The identified compounds were quantified by comparing the peak area of individual compounds with the peak area of the standard used with a concentration of 5 mg/mL.

3.8. Quality Analysis of M. koenigii Leaves

3.8.1. Colour Analysis

The loss of the colours in the leaves may cause a loss in the bioactive compounds, given that the colour of plant leaves is correlated to the bioactive compounds contributed to the pharmacological

properties [41]. Therefore, the colour analysis was conducted in the current study to evaluate the correlation between the colour parameters of the dried leaves and the antioxidant activity. The colour of *M. koenigii* leaves dried via CD, MVD, CPD-MVFD, and FD were determined following the procedure described by Chua et al. [23]. The *M. koenigii* leaves after the drying process were grounded into powder form. The colours were determined with a Minolta a Chroma Meter CR-200 (Minolta Co. Ltd., Osaka, Japan). Colour data were expressed in L^*, a^* and b^*. Each measurement was carried out five times, and the mean value was obtained.

3.8.2. Water Activity Analysis

Water activity corresponds to the amount of water available for the degradation reactions of antioxidant activity caused by the microorganisms [42]. Hence, the water analysis of dried *M. koenigii* leaves for CD, MVD, CPD-MVFD, and FD was conducted following the procedure described by Chua et al. [20] to evaluate the stability of dried *M. koenigii* leaves. The water content of dried leaves was determined using a water activity meter (Aqualab 4TE, Pullman, WA, USA). Powdered leaves were placed and spread up across the sample cup and inserted into the measuring chamber. The average temperature of the measuring chamber was 24.9 ± 0.05 °C.

3.9. Statistical Analysis

Results were expressed as mean ± standard deviation. The error bars in the figures indicated the standard deviation. Differences between means were analysed using one-way ANOVA through SPSS 23 (IBM, New York, NY, USA). Significant differences ($p \leq 0.05$) between means were determined using Tukey's test. The thin-layer modelling of CD, MVD and CPD-MVFD was performed using Table Curve 2D windows v2.03 (Jandel Scientific Software, San Jose, CA, USA), and goodness of fit was evaluated using R^2, RMSE and χ^2. The correlation analysis was determined using the Pearson correlation test through SPSS 23 (IBM, New York, NY, USA) [17].

4. Conclusions

The energy usage in the current drying process of herbal plants is a major concern as the commercial drying method consumes up to 60% of the total energy. Hence, the hybrid drying methods were proposed to study the energy consumption in comparison to the energy usage of the convective hot-air drying (CD). Both hybrid drying methods (MVD and CPD-MVFD) effectively reduced the specific energy consumption. Notably, MVD as the finishing drying method reduced the specific energy consumption by 67.3% (kJ/g fw) and 48.9% (kJ/g water) in comparison to CD at 50 °C. In the drying modelling of CD, MVD and CPD-MVFD, the modified Page model demonstrated good fit to the empirical data obtained. FD showed a promising antioxidant capacity result and MVD at 6 W/g retained a promising amount of TPC. Retained TPC was contributed to the antioxidant capacity (ABTS and FRAP) in *M. koenigii* leaves. β-phellandrene (2.647 mg/g dw), α-pinene (1.711 mg/g dw) and sabinene (1.374 mg/g dw) were identified as the major volatile compounds in dried *M. koenigii* leaves. Colour analysis showed MVD's high performance in preserving the colour parameters of *M. koenigii* leaves under all conditions. In the case of water activity, all drying methods showed that the sample conditions were shelf-stable. Furthermore, the brightness of the dried *M. koenigii* leaves was correlated to the TPC with a Pearson coefficient of 0.880 ($p \leq 0.01$). However, further investigation on the correlations of the single pigments to the antioxidant capacity and TPC are required. In summary, MVD showed a promising reduction in energy consumption as well as high recovery in TPC and volatile compounds.

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