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Phenolic compounds and antioxidant activity are differentially affected by drying processes in celery, coriander and parsley leaves

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Summary Convective drying (CO) is the most common technique for drying herbs, although it may diminish phenolic compounds content. Microwave drying (MW) has been suggested as an alternative, although its effects on phenolic compounds remain to be explored. The aim of this study is to compare the effects of different convective temperatures (40, 80 and 120 °C) and microwave powers (100, 500 and 1000 W) on drying time, polyphenols classes (HPLC-DAD) and antioxidant capacity in celery, coriander and parsley leaves. Microwave procedure reduced drying time (MW: 3 h–7 min; CO: 25–1.5 h) resulting more effective than CO. Increasing temperatures led to an initial decrease in total phenolic content (TPC) and antioxidant activity followed by an increase, potentially by the release of bound phenolic acids. Regarding MW, the highest polyphenol content was generally observed at 500 W, probably due to the hydroxycinnamic acid's rise. Antioxidant capacity was similar at 500 and 1000 W. There was a good agreement between TPC and antioxidant capacity. Consequently, MW seems a good alternative to reduce drying time. However, the maintenance or improvement of phenolic activity depends on the food matrix. Indeed, the best drying system and conditions to preserve phenolic compounds were variable depending on the herb type: celery (CO at 40 °C), coriander (both CO at 40 °C and MW at 500 W) and parsley (MW at 100 W).

Keywords Antioxidant activity, convective drying, culinary herbs, microwave drying, phenolic compounds.

Introduction

Culinary herbs and spices contribute significantly to the supply of polyphenols in the diet and are responsible for their antioxidant and anti-inflammatory effects (Deekshith *et al.*, 2021). They may also improve the sensory quality of food as well as add nutritional and functional value to food products (Sekhon *et al.*, 2016). Hence, food industry and consumers have high expectations regarding the quality of these aromatic (and often colourful) ingredients to refine foods and dishes (Schaarschmidt, 2016).

Celery (*Apium graveolens* L.), coriander (*Coriandrum sativum* L.) and parsley (*Petroselinum crispum* L.), annual herbs of the family Apaceae or Umbelliferae, are popular herbs widely used in many regions and find applications as culinary herbs or as minor additives in salads (fresh herbs) and herbal teas (dry leaves/stalks), as well as flavouring agents in the traditional food, functional foods, nutraceuticals, pharmaceutical and cosmetic industries (Kamel, 2013). Nevertheless, it is known that fresh produce agri-food supply chains are characterised by the perishability of the food product, seasonality, long supply lead time and uncertainty in harvest yield (Onggo *et al.*, 2019). Therefore, being drying is one of the primary preservation methods for the products (Hnin *et al.*, 2021). It is also the most common preservation method used to ensure safety against microbial attacks and control

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quality during storage, thereby making crops available all year long (Ali *et al*., 2020). Its goals include to preserve foods and increase their shelf life by reducing the water content and water activity; to avoid the need for use of refrigeration systems for transport and storage, which are expensive; to reduce space requirements for storage and transport and to diversify the supply of foods with different flavours and textures, thus offering the consumers a great choice when buying foods (Guiné, 2018).

Different drying methods and drying conditions, including time, temperature, microwave power level and air velocity, have link with various energy consumption and significant effects on phytochemicals and antioxidant properties of the samples (Saifullah *et al*., 2019). Convective drying (CO), also referred as hot-air or conventional drying, is the most economical and widely adopted technique in the food industry, although it requires long drying times and high air temperatures (Karam *et al*., 2016). However, in this type of drying, the retention of bioactive substances is poor due to high temperatures, long time and the presence of oxygen (Siebert *et al*., 2018; Jia *et al*., 2019). Therefore, microwave technology is widely used in drying process due to its rapid heat transfer, volumetric heating, pollution-free and selective heating characteristics (Su *et al*., 2020). Microwave drying (MW), which, in the last decade, has been widely studied to treat not only liquid samples, but also various kinds of solid materials, including food, vegetables, fruits, woody materials and ores, is currently attracting researchers' interest as an alternative non-conventional heating source for biomass and wastes processing, due to the advantages associated with dielectric heating effects (Miccio *et al*., 2020). Compared to the existing drying techniques, microwave drying method comes to the fore for saving time and energy as well as enabling the production of high-quality final products (Keser *et al*., 2020) and when compared with the conventional hot-air drying method, microwave drying is more rapid, uniform and energy efficient (Liu *et al*., 2020).

The increasing demand for high-quality shelf-stable dried products requires the design, simulation and further optimisation of the drying process with the purpose of accomplishing not only the efficiency of the process but also the final quality of the dried product (Hihat *et al*., 2017). For that reason, it is important to investigate in detail how thermal processing affects the contents of phytochemicals and antioxidant activities. Thus, although a previous study compared oven and microwave effect on coriander leaves (Hihat *et al*., 2017), a systematic comparison between convective and microwave drying in coriander, celery and parsley has not been performed. Therefore, this study aimed to evaluate whether the use of three different conditions for the two drying techniques led to substantial modifications in polyphenol content and activity in the three herbs.

Materials and methods

Plant material

The study was carried out on leaves of celery (*Apium graveolens* L.), coriander (*Coriandrum sativum* L.) and parsley (*Petroselinum crispum* L.) purchased from the local market in the region of Bejaia, Algeria. The leaves were separated from the stems. After sorting, the samples were packed in glass containers and refrigerated (4 ± 2 °C) for a maximum of 2–3 days. Before being dried, celery, parsley and coriander leaves were washed with water. For washing, abundant tap water was used directly to remove dust and impurities. As for the removal of moisture after washing, the salad spinner was used to remove most of the moisture, after which the samples were spread out on absorbent paper and given time to remove the rest.

Chemicals

Ethanol and other basic reagents were obtained from Panreac (Barcelona, Spain). Folin–Ciocalteu's phenol reagent, acetic anhydride, methanol, acetone and diethyl ether were obtained from Scharlab (Sentmenat, Spain). Gallic acid, 1-methylimidazole, 2,2-Diphenyl-1-picrylhydrazyl (DPPH), 6-hydroxy-2,5,7, 8-tetramethylchroman-2-carboxylic acid (Trolox) and 2,4,6-tri (2-pyridyl)-s-triazine (TPTZ) were from Sigma-Aldrich Chemical (St. Louis, MO, USA). All chemicals used were of analytical grade.

Drying processes

Two drying processes were used: CO at three different temperatures (40, 80 and 120 °C) in a NUVE type FN 400 ventilated oven and MW at three different powers (100, 500 and 1000 W) using a domestic microwave (Maxipower, model MASMO23S). During the drying process, 100 g of fresh leaves from different matrices was spread out on a plate and dried. Three replicates were carried out for each of the applied drying conditions. The samples were weighed periodically using a digital scale with an accuracy of 0.01 g (RADWAG, WPS 600/C/2) until the weight was stable, so that the drying kinetics can be plotted in terms of loss of mass as a function of the drying time. The drying time is thus the average of the times of the three repetitions needed to reach weight stability. The samples dried by both processes were then ground using an electric mill and the powders obtained were stored hermetically sealed in glass vials protected from light and moisture.

Antioxidants in dried powders

Extraction and determination of phenolic compounds

The method used for the extraction of phenolic compounds was previously described by Pérez-Jiménez *et al.* (2008). The procedure consisted of extracting polyphenols from the samples by stirring constantly at room temperature with methanol/water (50:50 v/v) and acetone/water (70:30 v/v). Then, the two supernatants were combined. In these extracts, the polyphenol content was quantified using two methods: the Folin–Ciocalteu assay (Singleton *et al.*, 1999), where results were measured spectrophotometrically at 750 nm on Multi-Modal reader microplates Synergy™ HTX; the Fast Blue method, which reacts more specifically with the hydroxyl groups of phenols (Lester *et al.*, 2012), measured at 420 nm (Lester *et al.*, 2012) on Multi-Modal reader microplates Synergy™ HTX. In both cases, gallic acid (0.01–0.28 mg mL^{-1}) was used as standard and the results were expressed as mg of gallic acid equivalents per g dry weight (mg GAE per g d.w).

Determination of in vitro antioxidant activity

The antioxidant activity estimation of the extracts of different dry powders was measured by the complementary methods: Ferric reducing antioxidant power; FRAP (based on reducing power) and DPPH (based on radical scavenging capacity).

FRAP

The antioxidant capacity of each sample was estimated according to the procedure described by Pulido *et al.* (2000) with some modifications. Briefly, 9 µL of test sample mixed with 26 µL of distilled water was mixed with 265 µL of FRAP reagent (prepared fresh and heated to 37 °C). After the incubation at 37 °C during 30 min, the absorption reading was taken at 595 nm on Multi-Modal reader microplates Synergy™ HTX. Trolox was used as standard and the results were expressed as mg of trolox equivalents per g dry weight (mg TE per g d.w).

DPPH

The DPPH radical-scavenging activity of the polyphenolic extracts was assayed with 20 µL of sample solution at the concentration of 5 mg mL^{-1} mixed with 280 µL of methanolic DPPH solution (100 µmol L^{-1}), following the method described by Huang *et al.* (2005). The mixture was shaken and kept at room temperature in the dark. Absorbance was measured at 517 nm on Multi-Modal reader microplates Synergy™ HTX.

HPLC-DAD

Phenolic compounds identification was carried out using an HPLC Agilent 1260 series system (Agilent Technologies, Santa Clara, CA, USA) equipped with a UV–Vis diode array detector (DAD). Samples were injected (210 µL, flow rate 0.41 mL min^{-1}) into a Gemini C18 Poroshell 120 EC-C18 column (4.6 mm × 2150 mm, particle size 2.75 µm) (Agilent Technologies) based on procedures previously reported. The gradient elution was carried out using water containing 0.1% trifluoracetic formic acid (Sigma Aldrich) as solvent A and acetonitrile containing 0.1% formic acid (Sigma Aldrich) as solvent B and it was applied as follows: 0 min, 45% B; 10 min, 17% B; 12 min, 17% B; 17 min, 25% B; 30 min, 50% B; 35 min, 100% B; 43 min, 1005% B; 50 min, 4% B; 55 min, 4% B. Following these conditions, a selection of widespread polyphenols belonging to different families was injected with two purposes: (i) potential identification in the samples; (ii) preparation of calibration curves that could be used for quantification/semi-quantification (in case that particular compound was not identified in the samples). These standards comprised four phenolic acids (gallic acid, ferulic acid, chlorogenic acid and p-coumaric acid) and five flavonoids (quercetin, kaempferol, quercetin 3-O-glucoside, kaempferol 3-O-rutinoside and luteolin). Maximum wavelengths for each polyphenol class were monitored: 280 nm for hydroxybenzoic acids, 320 nm for hydroxycinnamic acids, 365 nm for flavonoids. Then, samples were injected, peaks with maximum at each wavelength were identified and area was obtained. In the case of quercetin 3-O-glucoside, this compound was identified in coriander, so a quantification based on the calibration curve of the standard (25–250 ppm) was performed. For the other signals, they did not correspond to the other injected standards, so semi-quantification with a representative compound in each polyphenol class was performed: gallic acid (5–50 ppm) for hydroxybenzoic acids, chlorogenic acid (10–200 ppm) for hydroxycinnamic acids, luteolin (0.1–50 ppm) for flavones and kaempferol 3-O-rutinoside (10–50 ppm) for flavonols. Concentration ranges for these standards were based on previous experience on their linearity range. It should be indicated that both flavones and flavanols exhibit their absorbance maximum at 365 nm; but, based on previous bibliography search, it was established that in the case of coriander the absorbance at this wavelength corresponded to flavonols, while in celery and parsley these compounds belonged to the flavone class.

Statistical analysis

Data normality was assessed by Kolmogorov–Smirnov test. When it was not met (drying time), data were log-transformed. Two-way ANOVA (factor: treatment and condition) was applied, followed by T3 Dunnett's *post hoc* analysis, due to lack of homogeneity of variances. Regarding conditions, those for heating and for microwaving where independently analysed. In the case of flavones in parsley where normality was not met even after log-transformation, Kruskal–Wallis test, followed by Mann–Whitney test when statistical differences were found, was applied. In all cases, significance level was fixed at 0.05. Data were processed using IBM SPSS Statistics software version 26. PCA was designed using the software (XLSTAT Version May 3, 2014).

Results and discussion

Drying kinetics

Variations in moisture content during the drying processes of celery, coriander and parsley leaves as a function of drying time are shown in Fig. 1, which represent CO and MW. In both treatments, it was observed that the slightest condition needed a much higher time for decreasing moisture content up to 10% wet basis (w.b) than the other two conditions. But the most remarkable observation was that MW was much more effective for drying samples than CO. For instance, in the case of celery convective drying at 40 °C required a total time of 1500 min (=25 h), while with microwave drying at 100 W the time was decreased to 177 min (≈3 h); the same tendencies were observed for the other samples (Table 1).

When drying time for reaching a moisture content of 10% w.b was compared (Table 1), significant differences ($P < 0.05$) were found between the processes in favour of MW, which significantly reduced the drying time compared to CO. As expected, significant differences ($P < 0.05$) were also observed within the same process, so that increasing the drying temperature leads to a significant reduction in drying time.

Overall, these results agree with those obtained with nettle leaves (Alibas, 2007) and rosemary leaves (Ali *et al*., 2020) as well as the drying of fruits and vegetables by different techniques (Arslan & Musa Özcan, 2010; Jia *et al*., 2019; Miccio *et al*., 2020), and the effect was attributed to the fact that the heating rate and efficiency of microwave heating strongly depend on the dielectric properties and the relaxation times of the reaction mixture, whereby the use of good microwave absorbing solvents results in very fast heating (Hoogenboom & Schubert, 2007).

Total phenolic content

Thermal processing is among the several factors which may influence the phenolic content of fruit and vegetables (Arkoub-Djermoune *et al*., 2016). Therefore, the current study investigated the effect of two different drying processes on the total polyphenol contents (TPC) on the three matrices, measured by two methodologies, Folin–Ciocalteu and Fast Blue assays (Table 2). No common tendency was observed for the different samples subjected to the drying processes. Thus, in the case of CO, in both celery and coriander a significant decrease ($P < 0.05$) was observed at 80 °C, followed by an increase at 120 °C. The decrease at 80 °C may be attributed to losses of the most unstable polyphenols, while the increase at 120 °C may have two possible explanations: (i) the formation of Maillard reaction products that interfere in polyphenol determination (Que *et al*., 2008; Vega-Gálvez *et al*., 2009), since it has reported that, temperature, drying time and pH of the materials in simulated conditions has been described as pertinent conditions for the Maillard reaction during the drying and processing (Li *et al*., 2020) or (ii) the release of higher molecular weight bound phenolic compounds (Bushra, 2012). Similar results have been obtained in previous studies (Vega-Gálvez, *et al*., 2009; Nguyen *et al*., 2015; Ali *et al*., 2020). But a different trend was observed for parsley, where no significant difference in TPC value was observed when the convective drying temperature increased.

In the case of MW, regardless of the type of matrix dried, it was found that the powders dried at 100 W had the lowest significant values ($P < 0.05$). Increases in content were also observed between the values obtained for powders dried at 500 and 1000 W. However, these increases were not as significant. The increase in polyphenol content with increasing microwave power from 100 to 500 W can be explained by the intense heat generated by microwaves, which create a high vapour pressure and temperature inside plant tissues, which increase with the intensification of the microwave power, thus disrupting the polymers in the plant cell walls. As a result, phenolic compounds from the cell wall or bound phenolic compounds may be released, resulting in the extraction of more phenolic compounds (Inchuen *et al*., 2010). Similar results were reported by Kubra & Jagan Mohan Rao (2012) for ginger and by Izli *et al*. (2017) for mango. The results obtained in this study, where increases up to 1000 W did not always mean a significant increase in polyphenol content suggest that 500 W was enough to modify the structure of the fibre matrix facilitating polyphenol extraction. In this sense, it was observed in dried orange leaves and peels the highest TPC increase at 450 W when testing a range from 100 to 850 W (Kammoun Bejar *et al*., 2011).

Polyphenol profile by HPLC-DAD analysis

Extracts from dried celery, coriander and parsley powders were analysed by HPLC-DAD to better understand the variations in phenolic components in these

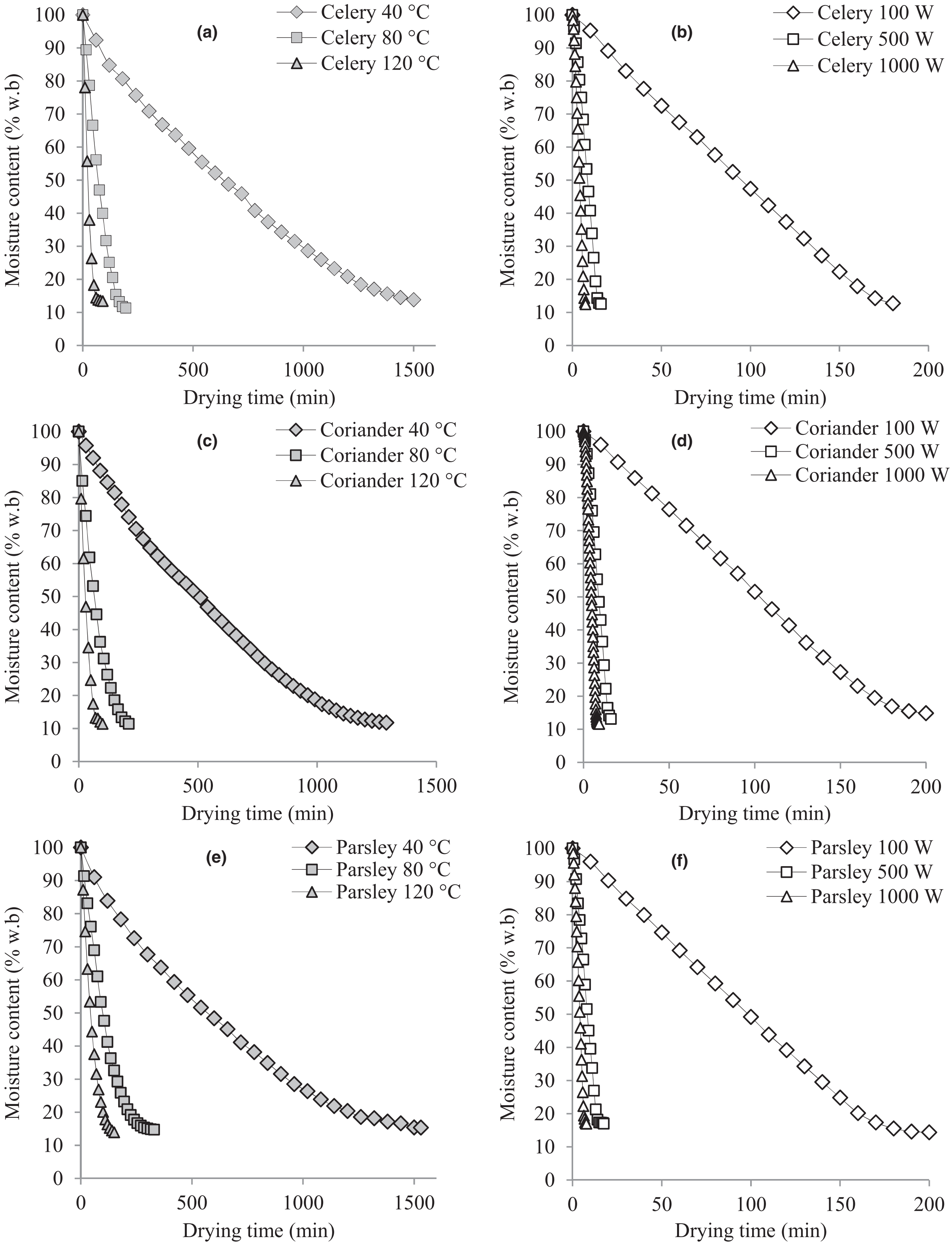

Figure 1 Changes in moisture contents of celery, coriander and parsley leaves vs. drying time during convective drying and microwave drying. Where (a): celery during convective drying, (b): celery during microwave drying, (c): coriander during convective drying, (d): coriander during microwave drying, (e): parsley during convective drying and (f): parsley during microwave drying.

Table 1 Drying times (min) for all samples subjected to drying treatments convective drying at different temperatures (°C) and microwave drying at different powers (W)

	Treatments					
	Convective drying			Microwave drying		
Samples	40 °C	80 °C	120 °C	100 W	500 W	1000 W
Celery	1500.00 ± 0.00^{a,A}	190.00 ± 8.66^{a,B}	86.67 ± 5.77^{a,C}	176.67 ± 5.77^{b,A}	15.33 ± 0.58^{b,B}	7.33 ± 0.00^{b,C}
Coriander	1280.00 ± 17.32^{a,A}	205.00 ± 8.66^{a,B}	100.00 ± 0.00^{a,C}	196.67 ± 5.77^{b,A}	16.00 ± 0.00^{b,B}	8.66 ± 0.29^{b,C}
Parsley	1510.00 ± 17.32^{a,A}	325.00 ± 8.66^{a,B}	150.00 ± 0.00^{a,C}	193.33 ± 5.77^{b,A}	17.17 ± 0.29^{b,B}	7.44 ± 0.19^{b,C}

Different lower case letters indicate significant differences between treatments ($P \leq 0.05$), while different capital letters indicate significant differences ($P \leq 0.05$) between conditions – those for heating and for microwaving were evaluated separately.

Table 2 Total phenolic content and antioxidant activity in spices subjected to different drying treatments

		Oven			Microwave		
Samples	Evaluated parameters	40 °C	80 °C	120 °C	100 W	500 W	1000 W
Celery	Folin–Ciocalteu (mg GAE per g d.w)	18.54 ± 2.56^{a,A}	12.53 ± 2.75^{a,C}	17.64 ± 3.66^{a,B}	11.10 ± 1.45^{b,C}	13.70 ± 0.95^{b,B}	13.99 ± 2.31^{b,A}
	Fast Blue (mg GAE per g d.w)	213.51 ± 14.77^{a,A}	175.91 ± 21.41^{a,A}	210.10 ± 15.13^{a,B}	183.77 ± 30.63^{a,B}	207.25 ± 26.11^{a,A}	209.04 ± 17.22^{a,A}
	FRAP (mg TE per g d.w)	308.54 ± 13.05^{a,A}	174.53 ± 15.85^{a,B}	286.44 ± 28.56^{a,A}	201.54 ± 15.25^{b,B}	212.42 ± 11.37^{b,B}	245.93 ± 19.33^{b,A}
	DPPH (%)	50.67 ± 3.70^{a,A}	30.17 ± 5.91^{a,B}	52.77 ± 2.72^{a,A}	26.19 ± 0.32^{b,B}	36.81 ± 4.79^{b,A,B}	37.48 ± 1.85^{b,A}
Coriander	Folin–Ciocalteu (mg GAE per g d.w)	21.23 ± 1.16^{a,A}	10.66 ± 0.49^{a,C}	16.57 ± 1.36^{a,B}	12.18 ± 1.15^{b,C}	20.87 ± 1.29^{b,A}	19.28 ± 0.58^{b,B}
	Fast Blue (mg GAE per g d.w)	271.25 ± 12.47^{a,A}	176.18 ± 15.05^{a,C}	215.62 ± 18.55^{a,B}	201.79 ± 23.06^{a,B}	237.43 ± 24.97^{a,A,B}	232.14 ± 12.62^{a,A}
	FRAP (mg TE per g d.w)	230.53 ± 18.04^{b,A}	123.33 ± 10.89^{b,C}	197.31 ± 19.79^{b,B}	193.22 ± 19.42^{a,C}	269.98 ± 22.41^{a,A}	235.43 ± 9.48^{a,B}
	DPPH (%)	38.60 ± 3.09^{b,A}	22.26 ± 2.21^{b,B}	39.23 ± 0.96^{b,A}	34.23 ± 1.65^{a,B}	47.73 ± 0.39^{a,A}	45.33 ± 4.73^{a,A,B}
Parsley	Folin–Ciocalteu (mg GAE per g d.w)	10.73 ± 1.27^{a,A}	12.22 ± 0.85^{a,A}	12.30 ± 1.81^{a,A}	10.32 ± 1.73^{a,B}	11.75 ± 1.27^{a,A,B}	13.63 ± 1.92^{a,A}
	Fast Blue (mg GAE per g d.w)	200.95 ± 14.36^{a,A,B}	212.80 ± 17.44^{a,A}	183.75 ± 16.43^{a,B}	218.00 ± 17.93^{b,A}	162.98 ± 15.35^{b,B}	178.94 ± 15.43^{b,B}
	FRAP (mg TE per g d.w)	97.88 ± 7.83^{b,A}	102.86 ± 4.74^{b,A}	97.36 ± 3.65^{b,A}	116.06 ± 6.57^{a,A}	114.77 ± 6.96^{a,A}	111.65 ± 11.60^{a,A}
	DPPH (%)	8.15 ± 1.27^{a,A}	9.32 ± 1.41^{a,A}	10.58 ± 1.65^{a,A}	7.91 ± 0.85^{b,A}	6.12 ± 0.58^{b,A}	6.32 ± 1.53^{b,A}

Table shows mean values with standard deviation of the mean. Different lower case letters indicate significant differences between treatments ($P \leq 0.05$), while different capital letters indicate significant differences ($P \leq 0.05$) between conditions – those for heating and for microwaving were evaluated separately. The DPPH (%) scavenging activity is for all samples at 5 mg mL^{-1}.

spices under different drying conditions, an aspect not previously explored. Thus, semi-quantification per classes was performed, excepting quercetin-3-O-gucoside, which could be identified in coriander (Table 3). The three samples exhibited hydroxybenzoic acids and hydroxycinnamic acids, while celery and parsley also contained flavones, and flavonols were detected in coriander. Previous studies on polyphenol characterisation in the samples of celery (Wolski *et al.*, 2002; Yildiz *et al.*, 2008; Yao & Ren, 2011), coriander (Nambiar *et al.*, 2010; Kaiser *et al.*, 2013) and parsley (Yildiz *et al.*, 2008) have highlighted the presence of hydroxybenzoic acids, hydroxycinnamic acids, flavones and flavanols in these samples.

In the three samples, hydroxycinnamic acids were the most abundant polyphenol class, so it is pertinent to evaluate specifically the modifications in these compounds due to drying. Thus, a similar tendency was observed for CO in the case of celery and coriander, with a significant ($P < 0.05$) decrease at 80 °C and a significant ($P < 0.05$) increase at 120 °C. This seems to indicate a first loss of certain hydroxycinnamic acids, followed by the release of cell wall bound phenolic compounds, also called non-extractable polyphenols, when the temperature increases; this has previously been observed, for instance, in cereal samples (Pérez-Jiménez & Saura-Calixto, 2005). Thus, between the two hypotheses suggested above for the variations in TPC values, that is, generation of Maillard reaction compounds or release of phenolic compounds from cell wall, the former seems to be more plausible. Nevertheless, this should be confirmed in a higher number of samples, since parsley showed a non-significant increase in hydroxycinnamic acids when oven temperature increased, what does not agree with TPC values. Also, for MW procedures, hydroxycinnamic acids in celery significantly ($P < 0.05$) increased with higher power values, while in coriander and parsley they did not show significant differences between conditions. As regards to the other polyphenol classes, flavonols and flavones tended to show a similar behaviour to that of hydroxycinnamic acids, while hydroxybenzoic acids showed an opposite one. Other studies have reported similar results, that is, drying conditions cause a decrease in a polyphenol class and an increase in another one, on lemon myrtle dried leaves (Saifullah *et al.*, 2019), on carrot (Keser, *et al.*, 2020) or on mango (Sarkar *et al.*, 2020), when testing different drying procedures.

In vitro antioxidant activity

The antioxidant activity of celery, coriander and parsley powders obtained after drying by two methods (CO and MW) was estimated by two antioxidant tests, FRAP and scavenging activity on DPPH (Table 2).

FRAP and DPPH methods showed tendencies quite similar to those obtained for TPC values and hydroxycinnamic acids content. For instance, in coriander dried with oven a significant ($P < 0.05$) decrease in antioxidant capacity measured by FRAP was observed from 40 to 80 °C, followed by a significant ($P < 0.05$) increase at 120 °C, nevertheless also significantly ($P < 0.05$) different to the initial value at 40 °C; this was the same behaviour found with Fast Blue assay.

The total polyphenol content and antioxidant capacity values agree with results reported by other

Table 3 Polyphenol content per classes (mg per 100 g dried product) by HPLC-DAD analysis in samples subjected to different drying treatments

		Oven			Microwave		
Samples	**Classes**	**40 °C**	**80 °C**	**120 °C**	**100 W**	**500 W**	**1000 W**
Celery	Hydroxybenzoic acids	148.09 ± 3.89^{a,B}	314.96 ± 28.18^{a,A}	71.32 ± 7.78^{a,C}	266.75 ± 42.38^{a,A}	115.92 ± 13.49^{a,B}	102.34 ± 5.43^{a,B}
	Hydroxycinnamic acids	2025.59 ± 218.89^{a,A}	33.02 ± 22.48^{a,C}	762.714 ± 49.46^{a,B}	670.87 ± 85.68^{b,C}	1442.07 ± 76.97^{b,B}	1876.2 ± 271.84^{b,A}
	Flavones	59.45 ± 7.98^{a,A}	17.08 ± 6.27^{a,B}	39 ± 5.44^{a,A}	29.89 ± 5.1^{b,A}	28.91 ± 1.41^{b,A}	33.44 ± 1.41^{b,A}
Coriander	Hydroxybenzoic acids	202.01 ± 8.85^{a,B}	443.13 ± 14.1^{a,A}	210.45 ± 12.35^{a,B}	289.81 ± 28.38^{b,A}	90.18 ± 5.42^{b,B}	86 ± 8.2^{b,B}
	Hydroxycinnamic acids	1540.95 ± 80.71^{a,A}	841.14 ± 1,98^{a,B}	1560.65 ± 105.43^{a,A}	1152 ± 102.7^{b,A}	1924.97 ± 80.95^{b,A}	1516.64 ± 100.95^{b,A}
	Quercetin glucoside	1470.04 ± 40.6^{a,A}	285.73 ± 11.47^{a,C}	474.38 ± 16.83^{a,B}	534.78 ± 91.84^{b,B}	838.14 ± 53.06^{b,A}	1096.92 ± 123.38^{b,A}
	Other flavonols	740.53 ± 46.93^{a,A}	245.37 ± 6.13^{a,C}	386.57 ± 24.09^{a,B}	495,99 ± 76,75^{b,B}	689.35 ± 47.72^{b,A}	534.18 ± 42.98^{b,A,B}
Parsley	Hydroxybenzoic acids	334.22 ± 25.83^{a,A}	996.04 ± 77.39^{a,B}	406.41 ± 73.21^{a,A}	923.26 ± 58.15^{a,A}	276.71 ± 49.40^{a,B}	293,05 ± 42.52^{a,B}
	Hydroxycinnamic acids	993.93 ± 211.59^{a,A}	1497.41 ± 41.81^{a,A}	1566.61 ± 23.91^{a,A}	1184.14 ± 161.60^{a,A}	1931.07 ± 573.70^{a,A}	1067.82 ± 103.39^{a,A}
	Flavones	n.d.^{a,B}	2.91 ± 0.08^{a,A}	n.d.^{a,B}	4.24 ± 0.49^{a,A}	n.d.^{a,B}	n.d.^{a,B}

Results provided as equivalents per class. except for quercetin glucoside. Different lower case letters indicate significant differences between treatments ($P \leq 0.05$), while different capital letters indicate significant differences ($P \leq 0.05$) between conditions – those for heating and for microwaving were evaluated separately.
n.d., non-detected.

authors, in several food matrices (Izli *et al*., 2017; Cherrat *et al*., 2019; Saifullah *et al*., 2019). The fact that similar tendencies were obtained when comparing different methodologies shows the consistency of the results.

Principal component analysis

For the purpose of comparison between CO and MW, the principal component analysis (data not shown) was designed to study the possible correlations existing between the different parameters (time, Folin–Ciocalteu, Fast Blue, FRAP, DPPH and the result of the characterisation) and to classify the different drying conditions (40, 80, 120 °C, 100, 500 and 1000 W).

The resulting PCA models explain 89.40%, 94.28% and 69.34% of the analysed data variability for celery, coriander and parsley respectively: while the component 2 (F2) of the PCA for celery and coriander represents 15.77% and 18.31% of the variance, respectively, and is mainly expressed by time and hydroxybenzoic acids, their component 1 (F1) represents 73. 63% and 75.98% of the variance and is expressed by the rest of the parameters, namely, by Folin–Ciocalteu, Fast Blue, FRAP, DPPH, hydroxycinnamic acids and flavones for celery and by Folin–Ciocalteu, Fast Blue, FRAP, DPPH, hydroxycinnamic acids, quercetin glucoside and other flavonols for coriander. As for parsley, its component 1 (F1) represents 41.63% of the variance and is mainly expressed by time, Fast Blue, DPPH, hydroxybenzoic acids and flavones and its component 2 (F2), which represents 27.70% of the variance, is represented by Folin–Ciocalteu, FRAP and hydroxycinnamic acids.

With the exception of parsley, this analysis allowed us to observe several correlations, such as between the two tests Folin–Ciocalteu and Fast Blue (r = 0.77 and r = 0.93), between the Folin–Ciocalteu test and the two antioxidant tests; FRAP (r = 0.93 and r = 0.89) and DPPH (r = 0.98 and r = 0.81), between the Fast Blue test and the two antioxidant tests; FRAP (r = 0.84 and r = 0.81) and DPPH (r = 0.81 and r = 0.70), as well as between the two antioxidant tests; FRAP and DPPH (r = 0.92 and r = 0.95) for celery and coriander respectively.

This analysis also allowed us to designate the best drying process for the three matrices (celery, coriander and parsley). Indeed, the ranking of the different drying conditions follows the same logic (trend) observed for the Fast Blue test, which allows us to deduce that CO at 40 °C is the best drying process for celery, CO at 40 °C and MW at 500 W are both better for coriander drying without any difference between them, and MW at 100 W is the best process for parsley drying.

Thus, the effectiveness of the treatment to preserve the polyphenol content seems to depend on the herb. Such a result is confirmed by the literature, as some authors have reported that MW was better in preserving TPC from different matrices (Inchuen *et al*., 2010; İzli, 2016; Izli *et al*., 2017; Sarkar *et al*., 2020) and others have reported that CO was better for others (Ali *et al*., 2020). The likely explanation for such results is that the phytochemical components of the different products changed differently, which may be caused by varying degrees of cell structure damage (Zhou *et al*., 2020), or even, in some cases, such as for total phenolics, there may be an increase or decrease due to thermal instability (Ratseewo *et al*., 2020).

Conclusion

This work focused on the comparative study of two drying processes, CO and MW, and their effect on polyphenol content and antioxidant activities of celery, coriander and parsley leaves. The results of the drying kinetics showed that the innovative microwave-assisted drying method allowed faster dehydration than the convective method. Overall, increasing temperatures led to an initial decrease in phenolic content followed by a further increase, potentially due to the release of previously bound phenolic acids. In the case of MW, the highest polyphenol content and antioxidant capacity were generally observed at a power of 500 W. Therefore, there was a good agreement between values of total polyphenol content and antioxidant capacity, while regarding HPLC-DAD analysis of polyphenol classes, they showed different tendencies depending on the conditions tested. Based on these results, the best drying system and conditions for drying foodstuffs in order to preserve phenolic compounds seems to depend on the type of sample.

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Conflict of interest

The authors report no declarations of interest.

Author contribution

Khokha Mouhoubi: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Lila Boulekbache-Makhlouf:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Khodir Madani:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Anastasia Palatzidi:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Jara Perez-Jimenez:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Inmaculada Mateos-Aparicio:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Alejandra Garcia-Alonso:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Methodology (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal).

Ethical approval

Ethics approval was not required for this research.

Peer review

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Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

References

Ali, A., Oon, C.C., Chua, B.L. *et al.* (2020). Volatile and polyphenol composition, anti-oxidant, anti-diabetic and anti-aging properties, and drying kinetics as affected by convective and hybrid vacuum microwave drying of *Rosmarinus officinalis* L. *Industrial Crops and Products*, **151**, 112463.

This reference is important because it consists in a comparative study of drying techniques on quality as our study, and supports our result that convective drying is better in preserving polyphenols (as in the case of celery).

Alibas, I. (2007). Energy consumption and colour characteristics of nettle leaves during microwave, vacuum and convective drying. *Biosystems Engineering*, **96**, 495–502.

Arkoub-Djermoune, L., Boulekbache-Makhlouf, L., Zeghichi-Hamri, S., Bellili, S., Boukhalfa, F. & Madani, K. (2016). Influence of the thermal processing on the physico-chemical properties and the antioxidant activity of a solanaceae vegetable: eggplant. *Journal of Food Quality*, **39**, 181–191.

Arslan, D. & Musa Özcan, M. (2010). Study the effect of sun, oven and microwave drying on quality of onion slices. *LWT – Food Science and Technology*, **43**, 1121–1127.

Bushra, S. (2012). Effect of drying techniques on the total phenolic contents and antioxidant activity of selected fruits. *Journal of Medicinal Plants Research*, **6**, 161–167.

Cherrat, S., Boulkebache-Makhlouf, L., Zeghichi, S. & Walker, G. (2019). Effect of different drying temperatures on the composition and antioxidant activity of ginger powder. *The Annals of the University Dunarea De Jos of Galati Fascicle VI – Food Technology*, **43**, 125–142.

Deekshith, C., Jois, M., Radcliffe, J. & Thomas, J. (2021). Effects of culinary herbs and spices on obesity: a systematic literature review of clinical trials. *Journal of Functional Foods*, **81**, 104449.

Guiné, R.P.F. (2018). The drying of foods and its effect on the physical-chemical, sensorial and nutritional properties. *International Journal of Food Engineering*, **4**, 93–100.

Hihat, S., Remini, H. & Madani, K. (2017). Effect of oven and microwave drying on phenolic compounds and antioxidant capacity of coriander leaves. *International Food Research Journal*, **24**, 503–509.

Hnin, K.K., Zhang, M., Ju, R. & Wang, B. (2021). A novel infrared pulse-spouted freeze drying on the drying kinetics, energy consumption and quality of edible rose flowers. *LWT*, **136**, 110318.

Hoogenboom, R. & Schubert, U.S. (2007). Microwave-assisted polymer synthesis: recent developments in a rapidly expanding field of research. *Macromolecular Rapid Communications*, **28**, 368–386.

Huang, D., Ou, B. & Prior, R.L. (2005). The chemistry behind antioxidant capacity assays. *Journal of Agriculture and Food Chemistry*, **53**, 1841–1856.

Inchuen, S., Narkrugsa, W. & Pornchaloempong, P. (2010). Effect of drying methods on chemical composition, color and antioxidant properties of Thai red curry powder. *Kasetsart Journal – Natural Science*, **44**, 142–151.

İzli, G. (2016). Total phenolics, antioxidant capacity, colour and drying characteristics of date fruit dried with different methods. *Food Science and Technology*, **37**, 139–147.

Izli, N., Izli, G. & Taskin, O. (2017). Influence of different drying techniques on drying parameters of mango. *Food Science and Technology*, **37**, 604–612.

It is important because it consists of a comparative study of drying techniques on quality as our study, and supports our result that microwave drying is better in preserving polyphenols (as the case of parsley).

Jia, Y., Khalifa, I., Hu, L. *et al.* (2019). Influence of three different drying techniques on persimmon chips' characteristics: a comparison study among hot-air, combined hot-air-microwave, and vacuum-freeze drying techniques. *Food and Bioproducts Processing*, **118**, 67–76.

The reference consists in a comparative study of drying techniques, and supports our result that the drying temperature has a significant effect on reducing the drying time.

Kaiser, A., Kammerer, D.R. & Carle, R. (2013). Impact of blanching on polyphenol stability and antioxidant capacity of innovative coriander (*Coriandrum sativum* L.) pastes. *Food Chemistry*, **140**, 332–339.

Kamel, S. (2013). Effect of microwave treatments on some bioactive compounds of parsley (*Petroselinum crispum*) and dill (*Anethum graveolens*) leaves. *Journal of Food Processing & Technology*, **4**, 4–6.

Kammoun Bejar, A., Kechaou, N. & Boudhrioua Mihoubi, N. (2011). Effect of microwave treatment on physical and functional properties of orange (*Citrus sinensis*) peel and leaves. *Journal of Food Processing & Technology*, **2**, 109.

Karam, M.C., Petit, J., Zimmer, D., Baudelaire Djantou, E. & Scher, J. (2016). Effects of drying and grinding in production of

fruit and vegetable powders: a review. *Journal of Food Engineering*, **188**, 32–49.
Keser, D., Guclu, G., Kelebek, H. *et al.* (2020). Characterization of aroma and phenolic composition of carrot (*Daucus carota* 'Nantes') powders obtained from intermittent microwave drying using GC–MS and LC–MS/MS. *Food and Bioproducts Processing*, **119**, 350–359.
Kubra, I.R. & Jagan Mohan Rao, L. (2012). Microwave drying of ginger (*Zingiber officinale* Roscoe) and its effects on polyphenolic content and antioxidant activity. *International Journal of Food Science & Technology*, **47**, 2311–2317.
Lester, G.E., Lewers, K.S., Medina, M.B. & Saftner, R.A. (2012). Comparative analysis of strawberry total phenolics via fast blue BB vs. Folin–Ciocalteu: assay interference by ascorbic acid. *Journal of Food Composition and Analysis*, **27**, 102–107.
Li, X., Gao, K., Jinfeng, B., Wu, X., Li, X. & Guo, C. (2020). Investigation of the effects of apple polyphenols on the chromatic values of weakly acidic lysine-fructose maillard system solutions. *LWT*, **125**, 109237.
This study provides an explanation for the increase in phenolic content observed when applying the 120°C drying temperature
Liu, L., Dai, X., Kang, H., Xu, Y. & Hao, W.(2020). Structural and functional properties of hydrolyzed/glycosylated ovalbumin under spray drying and microwave freeze drying. *Food Science and Human Wellness*, **9**, 80–87.
Miccio, M., Pierri, R., Cuccurullo, G., Metallo, A. & Brachi, P. (2020). Process intensification of tomato residues drying by microwave heating: experiments and simulation. *Chemical Engineering and Processing – Process Intensification*, **156**, 108082.
Nambiar, V.S., Daniel, M. & Guin, P. (2010). Characterization of polyphenols from coriander leaves (*Coriandrum sativum*), red amaranthus (*A. paniculatus*) and green amaranthus (*A. frumentaceus*) using paper chromatography and their health implications. *Journal of Herbal Medicine and Toxicology*, **4**, 173–177.
Nguyen, V.T., Van Vuong, Q., Bowyer, M.C., Van Altena, I.A. & Scarlett, C.J. (2015). Effects of different drying methods on bioactive compound yield and antioxidant capacity of *Phyllanthus amarus*. *Drying Technology*, **33**, 1006–1017.
Onggo, B.S., Panadero, J., Corlu, C.G. & Juan, A.A. (2019). Agri-food supply chains with stochastic demands: a multi-period inventory routing problem with perishable products. *Simulation Modelling Practice and Theory*, **97**, 101970.
Pérez-Jiménez, J., Arranz, S., Tabernero, M. *et al.* (2008). Updated methodology to determine antioxidant capacity in plant foods, oils and beverages: extraction, measurement and expression of results. *Food Research International*, **41**, 274–285.
Pérez-Jiménez, J. & Saura-Calixto, F. (2005). Literature data may underestimate the actual antioxidant capacity of cereals. *Journal of Agriculture and Food Chemistry*, **53**, 5036–5040.
Pulido, R., Bravo, L. & Saura-Calixto, F. (2000). Antioxidant activity of dietary polyphenols as determined by a modified ferric reducing/antioxidant power assay. *Journal of Agriculture and Food Chemistry*, **48**, 3396–3402.
Que, F., Mao, L., Fang, X. & Wu, T. (2008). Comparison of hot air-drying and freeze-drying on the physicochemical properties and antioxidant activities of pumpkin (*Cucurbita moschata* Duch.) flours. *International Journal of Food Science & Technology*, **43**, 1195–1201.
Ratseewo, J., Meeso, N. & Siriamornpun, S. (2020). Changes in amino acids and bioactive compounds of pigmented rice as affected by far-infrared radiation and hot air drying. *Food Chemistry*, **306**, 125644.
Saifullah, M., McCullum, R., McCluskey, A. & Vuong, Q. (2019). Effects of different drying methods on extractable phenolic compounds and antioxidant properties from lemon myrtle dried leaves. *Heliyon*, **5**, e03044.
Sarkar, T., Salauddin, M., Hazra, S.K. & Chakraborty, R. (2020). Effect of cutting edge drying technology on the physicochemical and bioactive components of mango (Langra variety) leather. *Journal of Agriculture and Food Research*, **2**, 100074.
Schaarschmidt, S. (2016). Public and private standards for dried culinary herbs and spices—Part I: standards defining the physical and chemical product quality and safety. *Food Control*, **70**, 339–349.
Sekhon, J.K., Maness, N.O. & Jones, C.L. (2016). Effect of compressed propane extraction on storage stability of dried cilantro (*Coriandrum sativum* L.). *Journal of Food Engineering*, **178**, 159–169.
Siebert, T., Gall, V., Karbstein, H.P. & Gaukel, V. (2018). Serial combination drying processes: a measure to improve quality of dried carrot disks and to reduce drying time. *Drying Technology*, **36**, 1578–1591.
Singleton, V.L., Orthofer, R. & Lamuela-Raventós, R.M.(1999). [14] Analysis of total phenols and other oxidation substrates and antioxidants by means of Folin-Ciocalteu reagent. In: *Methods in enzymology*. Vol. **299**, Pp. 152–178. Amsterdam: Elsevier.
Su, D., Lv, W., Wang, Y., Wang, L. & Li, D. (2020). Influence of microwave hot-air flow rolling dry-blanching on microstructure, water migration and quality of pleurotus eryngii during hot-air drying. *Food Control*, **114**, 107228.
Vega-Gálvez, A., Di Scala, K., Rodríguez, K. *et al.* (2009). Effect of air-drying temperature on physico-chemical properties, antioxidant capacity, colour and total phenolic content of red pepper (*Capsicum annuum*, L. var. Hungarian). *Food Chemistry*, **117**, 647–653.
Wolski, T., Dyduch, J. & Najda, A. (2002). Evaluation of content and composition of phenolic acids and tannins in leaf dry matter of two celery cultivars (*Apium graveolens* L. var. Dulce Mill. Pers.). *Electronic Journal of Polish Agricultural Universities (EJPAU)*, **5**.
Yao, Y. & Ren, G. (2011). Effect of thermal treatment on phenolic composition and antioxidant activities of two celery cultivars. *LWT – Food Science and Technology*, **44**, 181–185.
Yildiz, L., Baskan, K.S., Tutem, E. & Apak, R. (2008). Combined HPLC-CUPRAC (cupric ion reducing antioxidant capacity) assay of parsley, celery leaves, and nettle. *Talanta*, **77**, 304–313.
Zhou, Y.-H., Vidyarthi, S.K., Zhong, C.-S. *et al.* (2020). Cold plasma enhances drying and color, rehydration ratio and polyphenols of wolfberry via microstructure and ultrastructure alteration. *LWT*, **134**, 110173.